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POWER REACTOR TECHNOLOGY

PREPARED BY GENERAL NUCLEAR ENGINEERING CORP. FOR THE U S ATOMIC ENERGY COMMISSION



FEBRUARY 1958

VOLUME 1

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POWER REACTOR TECHNOLOGY

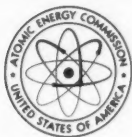
a review of recent developments *prepared by*

GENERAL NUCLEAR ENGINEERING CORP.

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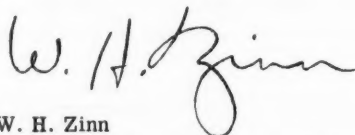


foreword

This quarterly review of reactor development has been prepared at the request of the Division of Information Services of the United States Atomic Energy Commission. Its purpose is to assist interested organizations in the task of keeping abreast of new results in reactor technology for civilian application.

The report is a concise discussion of selected phases of research and development for which there have been significant advances or a heightened interest in the past few months. It is not meant to be a comprehensive abstract of all material published during the quarter, nor is it meant to be a treatise on any part of the subject. The intention is to cover the various areas of reactor development from the general viewpoint of the reactor designer rather than from the more detailed points of view of specialists in the individual areas. However, papers which are thought to be of particular significance or particular usefulness in specialized fields will be mentioned in short notes. In the over-all plan of the report, it is intended that various subjects will be treated from time to time and will be brought up to date at that time.

Any interpretation of results which is given represents only the opinion of the editors of the report, who are General Nuclear Engineering Corporation personnel. Readers are urged to consult the original references wherever possible in order to obtain all the background of the work reported and to obtain the interpretation of the results given by the original authors.



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P.D.
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vol
no. 2

contents

iii

Foreword

General Research and Development

- | | | |
|----|-----|--|
| 1 | I | Reactor Physics |
| 16 | II | Reactor Dynamics and Safety |
| 18 | III | Fuel Elements |
| 34 | IV | Other Reactor Materials |
| 42 | V | Fuel Cycles: Plutonium Recycle |
| 44 | VI | Fluid Fuels: UO_2 -NaK Slurry |
| 45 | VII | Components |

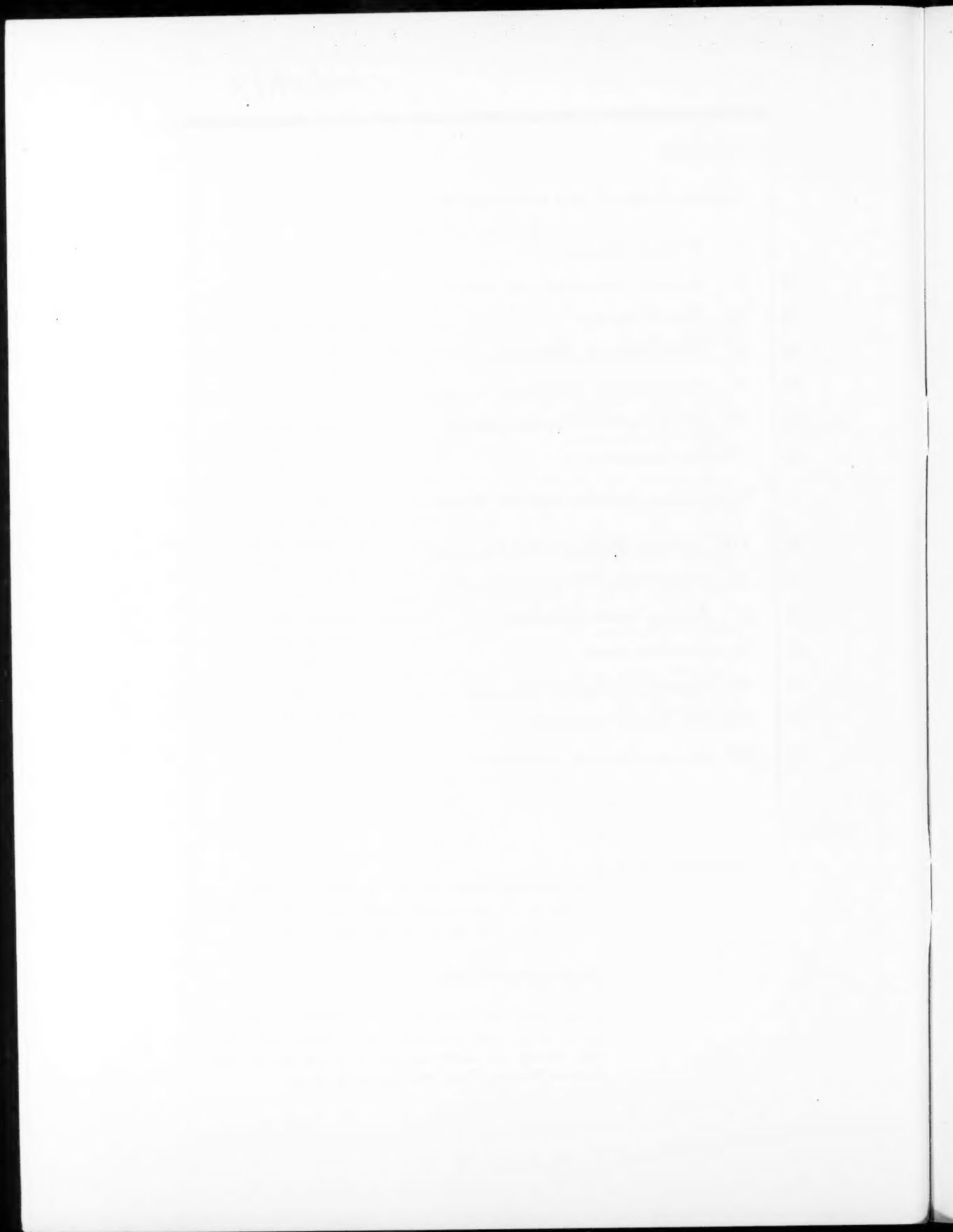
Progress on Specific Reactor Types

- | | | |
|----|------|------------------------------|
| 46 | VIII | Aqueous Homogeneous Reactors |
| 62 | IX | Pressurized Water Reactors |
| 64 | X | Boiling Water Reactors |
| 66 | XI | Fast Reactors |
| 68 | XII | Organic Moderated Reactors |
| 68 | XIII | Gas Cooled Reactors |
| 69 | XIV | Molten Fluoride Reactors |

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Although fairly extensive measurements on multiplying lattices which use simple solid fuel rod configurations have been made and reported in the past (the series of volumes covering the International Conference on the Peaceful Uses of Atomic Energy in Geneva, 1955, contain good summaries of these measurements), there has been little information on lattices which employ subdivided or clumped fuel elements. Information of the latter type is of great importance because it is with such fuel elements that the high specific powers favorable to economic power generation can be attained. It therefore appears worth while to summarize those more recent buckling measurements on clumped elements which have general applicability. Some measurements have been made in D_2O (references 1, 2, 4, and 5), mixtures of D_2O and H_2O (reference 3), and graphite.¹¹

Recent analyses of some of the D_2O data indicate⁶ that rather good theoretical results can be obtained when P-3 calculation of the thermal-flux distribution is combined with the Spinrad method for the fast fission factor and the method of Critch for the resonance escape probability.⁷

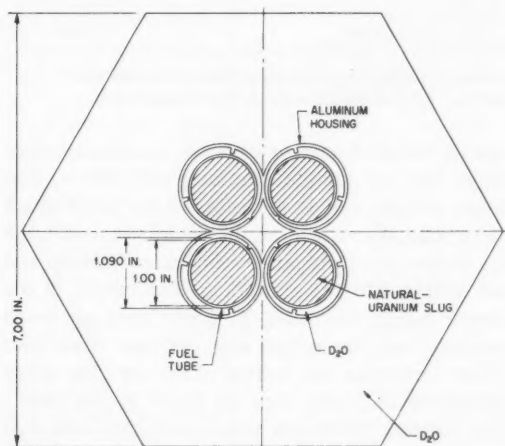


Figure 1—Fuel cluster in unit cell of D_2O lattice.

A further aspect of the problem of lattice theory has to do with the neutron energy spectrum in the thermal range. Questions relating to the thermal spectrum are perhaps most important in the relatively "black" lattices characteristic of H_2O moderated reactors. When plutonium becomes an important constituent of the fuel, however, because of either long fuel element lifetime or plutonium recycling, these questions may become important for any lattice type. The recent advances in the understanding of neutron thermalization and the thermal energy spectrum are summarized on pp. 5-9.

Other subjects covered in this section are the resonance escape probability and the stability of the flux distribution in reactors having very high thermal neutron flux.

D_2O Moderated Lattices

The lattice for all the D_2O measurements is a triangular one with equilateral fuel element spacing of 7 in. Thus each fuel assembly, consisting of a cluster of fuel slugs or fuel plates, lies in the center of a hexagonal cell which is 7 in. across flats (Fig. 1).

Bucklings were measured for lattices consisting of clusters of four natural-uranium fuel slugs in D_2O - H_2O mixtures of several proportions.³ The measurements were made on an exponential assembly roughly 5 ft in diameter by 5 ft high. The fuel slugs were 1.00 in. in diameter and 8.07 in. long; aluminum spacers in the form of disks 0.330 in. thick were used between the ends of adjacent slugs. The volume fractions of materials are given in Table I-2, and the measured values of buckling for the various D_2O - H_2O mixtures are given in Table I-1. The thermal utilization was determined for three of the cases by making detailed thermal-flux traverses. The pertinent quantities are listed in Table I-2. The other lattice constants were evaluated as follows. It was assumed that the measured value of τ for the appropriate D_2O -

H₂O mixture (see pp. 4-5) applied for the assembly in each case. η was taken as 1.34 and ϵ , from previous measurements, as 1.027. The

Table I-1 BUCKLING AS FUNCTION OF PERCENTAGE OF D₂O IN D₂O-H₂O MIXTURE³

D ₂ O, %	Material buckling, μB^*
99.8	558
98.7	541
96.3	503
95.4	476
94.0	403
91.8	275

*1 $\mu B = 10^{-6} \text{ cm}^{-2}$.

diffusion constant D was approximated in terms of V (the volume fraction of D₂O or H₂O) by

$$\frac{1}{D} = \frac{V_{D_2O}}{D_{D_2O}} + \frac{V_{H_2O}}{D_{H_2O}}$$

Table I-2 RESULTS OF DETAILED FLUX TRAVERSES³
(Moderator purity, mole % D₂O)

Component	Σ_a , cm^{-1}	Lattice constants for 99.8% D ₂ O			Lattice constants for 98.4% D ₂ O			Lattice constants for 91.8% D ₂ O		
		V	$\bar{\phi}$	$\Sigma_a V \frac{\bar{\phi}}{\bar{\phi}_{\text{cell}}}$	V	$\bar{\phi}$	$\Sigma_a V \frac{\bar{\phi}}{\bar{\phi}_{\text{cell}}}$	V	$\bar{\phi}$	$\Sigma_a V \frac{\bar{\phi}}{\bar{\phi}_{\text{cell}}}$
Fuel	0.3187	0.07403	37.0	0.01074	0.07403	36.8	0.01054	0.07403	36.2	0.01033
Aluminum	0.0121	0.02801	45.9	0.000191	0.02801	47.8	0.000197	0.02801	46.8	0.000191
D ₂ O	2.71×10^{-5}	0.8961	86.1	0.000026	0.8835	87.4	0.000025	0.8243	87.7	0.000024
H ₂ O	0.0192	0.001796	86.1	0.000037	0.01437	87.4	0.000293	0.07363	87.7	0.00150
Unit cell		1.000	81.3	0.01099	1.000	82.3	0.01106	1.000	82.6	0.01204
Thermal utilization, f				0.577			0.953			0.858

* V is volume fraction; $\bar{\phi}$ is average flux in the component in arbitrary units; $\bar{\phi}_{\text{cell}}$ is average flux in the unit cell in arbitrary units; Σ_a is macroscopic absorption cross section for thermal neutrons averaged over a Maxwellian distribution.

The diffusion constant, along with the effective absorption cross section obtained in evaluating the thermal utilization, gave an evaluation of the diffusion area L^2 . Using these values for the lattice constants, and the measured material buckling, the resonance escape probability p can be determined from the equation for the material buckling B^2

$$p = \frac{1 + M^2 B^2}{\eta \epsilon f}$$

It was shown that a reasonable approximation for the variation of p with moderator composition is

$$p = e^{-\alpha/(\xi \Sigma_s)_m}$$

where $(\xi \Sigma_s)_m$ is the slowing-down power of the mixture = 1.390-1.213 (mole fraction D₂O in mixture) and α (= 0.0345) is obtained from the value of p for this lattice for 100 per cent D₂O. For 92 per cent D₂O the relation gives $p = 0.882$, whereas the value from the buckling measurement is 0.878 (the value of p for 100 per cent D₂O is 0.823).

Hollow slugs of natural uranium were used in a lattice similar to that of Fig. 1, with D₂O of high purity as the moderator. Buckling measurements were made on a small lattice of this type, which was surrounded by a larger lattice of slightly different composition, the combined assembly being critical. The material buckling of the experimental part of the lattice was evaluated in terms of the critical moderator height, the method having been previously calibrated by means of lattice sections of known buckling.

Slugs of two sizes were used: the larger was hollow, having an outside diameter of 1.048 in.

and an inside diameter of 0.471 in.; the smaller slugs had an outside diameter of 1.000 in. (the same as the outside diameter of the solid slugs described above) and an inside diameter of 0.550 in. Slugs of both types were jacketed inside and out with 0.030 in. aluminum. The lattice of the larger slugs contained 87.5 per cent as much uranium as the solid-slug lattices described above, whereas the lattice of the smaller slugs contained 69.1 per cent as much as the solid-slug lattice. Bucklings were measured with D₂O inside the hollow slugs and in the annulus between the slug and the aluminum housing (wet) and without D₂O in these two volumes (dry). The measured bucklings are compared with those for solid-slug lattices in Table I-3.

The measured average fluxes and other quantities which determine f are given, for the case of the large hollow slugs, in Table I-4, and the lattice parameters for the same case are com-

Table I-3 D₂O LATTICES;
BUCKLINGS WITH HOLLOW AND SOLID SLUGS⁴

Slug type	Buckling (B^2), ΔB^2 wet to dry,	
	μB	μB
Large, hollow	481	+11
Small, hollow	496	
Solid (1 in. diameter)	519	-12

pared with those for the solid-slug case in Table I-5. L^2 is computed from the data of Table I-4, using the listed value of D (Table I-5). The product $p\epsilon$ is then computed from the equation for the material buckling. The occurrence of nearly identical values for $p\epsilon$ is fortuitous, the change in effective surface-mass ratio being balanced by the higher moderator-uranium ratio for the hollow slugs.

Table I-4 D₂O LATTICES;
EFFECTIVE ABSORPTION CROSS SECTIONS⁴
(Large Hollow-slug Lattice)

Material	$\bar{\phi}_{th}$	d_i^*	$\frac{V_i}{V_{cell}}$	Σ_a	$\frac{\Sigma_a d_i V_i}{V_{cell}}$
Uranium	42.5	0.520	0.0648	0.319	0.01075
Aluminum	50.7	0.620	0.0362	0.0121	0.00027
Moderator	85.9	1.050	0.899	0.0001	0.00009
Cell	81.8				0.01111

$$*d_i = \frac{\bar{\phi}_{th} \cdot i}{\bar{\phi}_{th} + \phi_{cell}}$$

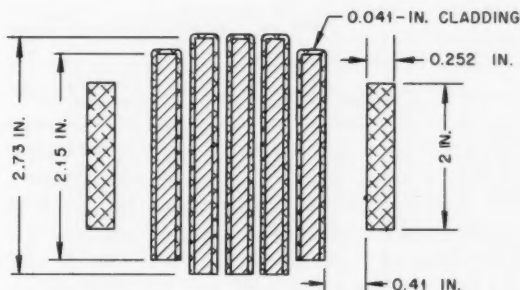
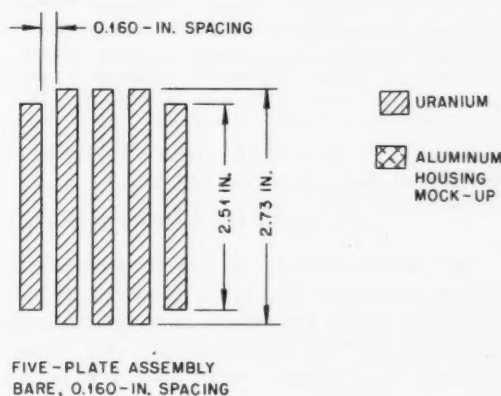
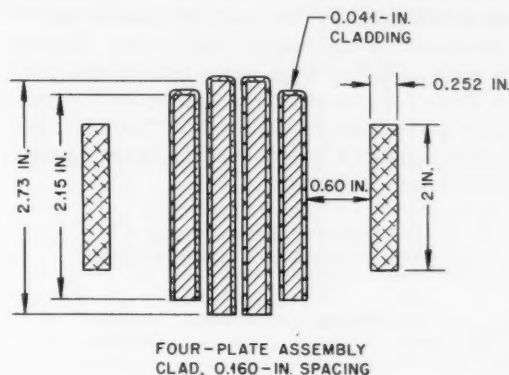
Table I-5 D₂O LATTICES;
LATTICE PARAMETERS FOR SOLID-SLUG
AND HOLLOW-SLUG LATTICES⁴

Parameters	Solid slugs	Large hollow slugs
η	1.33	1.33
f	0.972	0.968
D	0.900 cm	0.900 cm
L^2	72.5 cm ²	81.0 cm ²
B^2	519 μB^*	481 μB^*
τ	127 cm ²	127 cm ²
M^2	200 cm ²	208 cm ²
k_∞	1.104	1.100
$p\epsilon$	0.854	0.855

$$*1 \mu B = 1 \text{ cm}^{-2} \times 10^{-6}$$

Small lattices made up of clusters of natural-uranium plates were investigated also by the technique of installing the small lattice as part of a larger critical one.⁵ The plates were all

0.180 in. thick, plated with 0.0008 nickel for corrosion protection. They were used with and without aluminum strips to simulate cladding materials and housing tubes. Figure 2 shows



FIVE-PLATE ASSEMBLY
CLAD, 0.160-IN. SPACING

Figure 2—Typical arrangements of fuel plates in D₂O lattices.

three typical plate arrangements. The lattice spacing is the same as that described above; the cluster of plates takes the place of the four fuel slugs of Fig. 1. Bucklings were measured, and the individual lattice constants were evaluated by the methods used in the investigation of the hollow slugs, above. Results of some of the buckling measurements are given in Table I-6, disadvantage factors for the five-plate arrangements are given in Table I-7, and lattice constants for some of the cases investigated are in Table I-8.

Table I-6 D₂O LATTICES;
BUCKLINGS FOR PLATE ASSEMBLIES⁵

No. plates	Spacing, in.	Cladding thickness, in.	Housing mock-up	B ² , μB
5	0.160	0.041	Present	405
5	0.160	0.041	Absent	553
5	0.160	0.0175	Absent	588
5	0.160	None	Absent	630
5	0.320	None	Absent	567
5	0.0	None	Absent	735
4	0.160	None	Absent	685
4	0.160	0.041	Present	475

Neutron Age in Mixtures of Light and Heavy Water

Age measurements of fission neutrons to indium resonance energy⁸ in mixtures of light and heavy water are useful in connection with the above buckling measurements. The measurements are summarized in Table I-9. During the same set of experiments, the age in 99.3 per cent D₂O containing 9.1 per cent of 2S aluminum

in the form of 1-in.-diameter parallel rods was measured. The age in the direction parallel to the rods was 140 cm², and in the direction perpendicular to the rods it was 108 cm², essentially the same as for the 99.3 per cent D₂O alone.

Table I-7 D₂O LATTICES;
DISADVANTAGE FACTORS FOR PLATE ASSEMBLIES⁵

	Plate spacing, in.		
	0	0.160	0.320
d _{Al} (0.041-in. cladding with housing)		0.652	
d _{Al} (0.041-in. cladding without housing)		0.538	
d _{Al} (0.0175-in. cladding without housing)		0.538	
d _U	0.458	0.502	0.592
d _{D₂O}	1.03	1.026	1.02
d _{Ni}	0.473	0.526	0.599

The preceding paragraph discussed neutron age measurements⁸⁻¹⁰ from a fission source in heavy water containing 1.2 to 8.2 mole % H₂O. Additional measurements have been made for a mixture containing 51.4 per cent H₂O. The age of fission neutrons to 1.4 ev was found to be 38.6 cm² as compared with 78 cm² and 109 cm² for 8.2 and 1.2 per cent H₂O, respectively. The curve of age vs. moderator purity shows that adding 1 per cent of H₂O to pure D₂O decreases the age by about 4 per cent. The rate of decrease in age diminishes as more H₂O is added. The authors of the referenced reports suggest that this tendency may make it advantageous to consider a reactor which uses about 75 mole % of D₂O because such a reactor could be small in size like an H₂O reactor and have neutron absorption characteristics not too drastically different from D₂O.

It has been shown previously that addition of small percentages of H₂O to small D₂O reactors

Table I-8 D₂O LATTICES; LATTICE CONSTANTS FOR PLATE ASSEMBLIES⁵

No. plates	Spacing, in.	Cladding thickness, in.	Housing mock-up	f	L ² , cm ²	k _∞	p
5	0.160	0.041	Present	0.942	93.5	1.089	0.847
5	0.160	0.041	Absent	0.968	96.1	1.123	0.850
5	0.160	0.0175	Absent	0.979	97.1	1.132	0.847
5	0.160	None	Absent	0.987	97.8	1.142	0.847
5	0.320	None	Absent	0.988	83.2	1.119	0.829
5	0.0	None	Absent	0.986	107.1	1.172	0.870
4	0.160	None	Absent	0.984	121	1.170	0.871
4	0.160	0.041	Present	0.939	115	1.115	0.870

*In all cases:

$$\begin{aligned} \Sigma_a \text{ aluminum} &= 0.012 \text{ cm}^{-1} & \phi &= 0.90 \text{ cm} \\ \Sigma_a \text{ moderator} &= 9.4 \times 10^{-6} \text{ cm}^{-1} & \tau &= 127 \text{ cm}^2 \\ \Sigma_a \text{ uranium} &= 0.322 \text{ cm}^{-1} & \eta &= 1.34 \\ \Sigma_a \text{ nickel} &= 0.346 \text{ cm}^{-1} & \epsilon &= 1.019 \text{ (estimated)} \end{aligned}$$

increases reactivity. Not much H_2O can be added without a serious decrease in conversion ratio however. The lower conversion ratio makes the loss in reactivity more rapid as irradiation of fuel progresses in power reactors. It has been suggested (The Babcock & Wilcox Co.) that by progressively adding H_2O to D_2O the reactivity may be increased as necessary to compensate for loss due to burn-out of fissionable material.

Graphite Lattices

Bucklings have been measured¹¹ in a small exponential lattice for enriched (1.007 wt. % U^{235}) hollow and solid cylindrical slugs. Solid slugs were 0.925 in. in diameter, and the hollow slugs had an outside diameter of 1.66 in. and an inside diameter of 0.94 in. Solid slugs of 1.66-in. diameter and nearly full density (approximately 1 per cent density loss) were obtained by inserting the small solid slugs in the larger hollow ones. The slugs were 4 in. long; aluminum disks 0.4 in. thick were inserted as spacers at 8-in. intervals. The 1.66-in. slugs were in aluminum cans of 0.028 in. wall thickness; the 0.925-in. slugs were in 0.035-in. aluminum cans. Bucklings for the 1.66-in.-diameter slugs at various spacings in a square lattice are given in Table I-10. The experimental error is stated to be about 10 μB .

Bucklings for the enriched 0.925-in. rods are given in Table I-11, along with bucklings for the same configuration with natural uranium. These slugs were contained in aluminum process tubes of radius 1.55 cm (see table for U-Al ratios).

Table I-9 AGE FROM FISSION ENERGY TO INDIUM RESONANCE IN MIXTURES⁸ OF D_2O AND H_2O

Slowing-down media, % D_2O	τ , cm ²	Estimated error, cm ²
99.8	109	± 3
99.0	107	± 5
98.4	106	± 3
95.3	93	± 3
94.0	86	± 3
91.8	78	± 3

Bucklings for the 1.66-in. hollow slugs are given in Table I-12. The "wet" lattice refers to the case in which the central hole of the slugs was partially filled (54 vol. %) with H_2O . The water was contained in aluminum tubes of volume 0.89 cm³/cm of slug length. The worth of this aluminum in terms of buckling was found

to be less than the precision of the measurements (its effect was measured to be $-3 \mu B$ in the $7\frac{1}{2}$ -in. lattice).

Bucklings for clusters of rods of 0.925 in. diameter in a square lattice of 14 in. spacing have been measured in an exponential assembly.

Table I-10 GRAPHITE LATTICES;
BUCKLINGS WITH 1.66-IN.-DIAMETER SOLID
ENRICHED SLUGS¹¹

Lattice spacing, in.	C/U (atom ratio)	Al/U (atom ratio)	Buckling, μB
$5\frac{3}{16}$	19.8	0.088	-157
$6\frac{3}{16}$	29.1	0.088	164
7	37.5	0.088	325
$7\frac{1}{2}$	43.4	0.088	367
$8\frac{3}{8}$	54.9	0.088	404
$10\frac{3}{8}$	85.2	0.088	370
$12\frac{3}{8}$	122.0	0.088	268

The rod clusters were contained in aluminum process tubes of 3.02 in. inside diameter and wall thickness 0.065 in. Bucklings for the dry and wet (process tube filled with H_2O) lattices are given in Table I-13.

Neutron Energy Spectra in the Thermal Energy Range

Significance

For the past two years, there has been a considerable amount of published work, both theoretical and experimental, concerning neutron spectra in the "thermal" energy range. The exact limitation on the thermal energy is a function of reactor composition, but broadly the term refers to neutrons whose energies are below a few electron volts. In general, the neutron spectrum can be divided into three energy regions: a slowing-down region where the neutron loses energy in each collision with the moderating atom, a thermal region in which the neutron can gain or lose energy in colliding with the moderating atom (thermalization), and a transition region in which both slowing down and thermalization are possible. The latter two energy regions are usually lumped together as the thermal energy range in reactor physics calculations.

For assemblies in which the neutron absorption is very small, the thermal-neutron flux is expected to possess a pure Maxwellian energy

distribution whose most probable energy is equal to kT , where k is the Boltzmann constant and T is the temperature of the moderator. The thermal flux is expected to join smoothly to a $1/E$ neutron slowing-down flux, or, as usually stated, the Maxwellian is expected to have a $1/E$

as one neutron energy group. One can only ascribe the proper averaging process for the neutron scattering, transport, and absorption properties by knowing the thermal spectrum, since all these processes are energy dependent. Although it is true that past calculations for highly

Table I-11 GRAPHITE LATTICES; BUCKLINGS WITH 0.925-IN.-DIAMETER ENRICHED- AND NATURAL-URANIUM SLUGS¹¹

Lattice spacing, in.	C/U (atom ratio)	Al/U (atom ratio)	H ₂ O/U (molar ratio)	Buckling, μB	
				1.007% U ²³⁵	Natural uranium
6 ³ / ₁₆	100.4	0.69	0	356	114.5
6 ³ / ₁₆ *	95.6	0.59	0	367	
7 ¹ / ₂	148.4	0.69	0	313	95.3
7 ¹ / ₂ (wet)†	148.4	0.69	0.174	297	66.4

*Continuous uranium rods; no aluminum spacers.

†Water between slugs and process tube.

tail. Such neutron flux energy distributions have been observed in experimental measurements of weakly absorbing assemblies. However, as neutron absorber is added, the thermal flux will be distorted from the pure Maxwellian. The physical explanation is that the lower energy neutrons have a shorter life expectancy in a region of higher absorption. The more rapid disappearance of these neutrons with increased absorption causes an upward shift of the most probable neutron energy. This effect is com-

absorbing assemblies have been reasonably successful in predicating fuel loading and reactivity changes without detailed consideration of thermal spectra, these calculations were usually limited to highly enriched reactors moderated by H₂O, in which thermal leakage is small and thermal utilization is insensitive to spectral changes. However, even for this type of reactor, inconsistencies have arisen for phenomena such as control-rod worth and water-gap peaking, which are functions of thermal

Table I-12 GRAPHITE LATTICES; BUCKLINGS FOR ENRICHED HOLLOW SLUGS (1.66 IN. OUTSIDE DIAMETER; 0.94 IN. INSIDE DIAMETER)¹¹

Lattice spacing, in.	Dry lattice			Wet lattice			
	C/U (atom ratio)	Al/U (atom ratio)	Buckling, μB	C/U (atom ratio)	Al/U (atom ratio)	H ₂ O/U (molar ratio)	Buckling, μB
6 ³ / ₁₆	42.3	0.25	253	42.3	0.25	0.18	306
7 ¹ / ₂	63.2	0.25	375	63.2	0.25	0.18	409
12 ³ / ₈	177.8	0.25	222	177.8	0.25	0.18	226

monly known as "spectrum hardening." Since the core region of a reactor will usually contain a considerable amount of absorber, the flux can deviate quite materially from the pure Maxwellian in practical cases. In fact, for the large fuel concentrations required in intermediate and fast assemblies, there is no longer any semblance of the Maxwellian distribution.

The necessity for knowing the detailed thermal-neutron spectrum becomes apparent when it is realized that in reactor calculations the entire thermal energy region is usually treated

diffusion length, because the thermal diffusion length is quite sensitive to the thermal-flux spectrum.

Since most critical experiments at present are performed near room temperature and at atmospheric pressure, whereas a power reactor in actual operation is at an elevated temperature and pressure, it is important to be able to predict the proper dependence of the thermal spectrum on moderator temperature and density. The spectral shift will not only affect the static behavior but also the kinetic behavior by changes

in the temperature coefficient and neutron generation time.

To illustrate the possible effect on reactor operation of spectral shift caused by temperature change in the moderator, one might consider a thermal reactor fueled with Pu^{239} . Pu^{239} has a large resonance at 0.3 eV which is in the previously defined transition energy region. Be-

ments to guide the theory. The experimental measurements are themselves rather complex in that a considerable amount of analysis must be performed to reduce the measured data to the desired form.

Several different methods for measuring spectra have been developed. For internal measurements foils and fission counters are used. In

Table I-13 GRAPHITE LATTICES; BUCKLINGS IN LATTICES COMPOSED OF CLUSTERS OF 0.925-IN.-DIAMETER NATURAL-URANIUM RODS¹¹

No. rods in cluster	Lattice spacing, in.	C/U (atom ratio)	$\text{H}_2\text{O}/\text{U}$ (molar ratio)	Al/U (atom ratio)	Buckling, μB	
					Wet	Dry
7	14	70.9	0.258	0.45	19	90
6	14	82.7	0.445	0.48	8	87
5	14	99.3	0.704	0.52	-69	83
5	10 $\frac{3}{8}$	52.0		0.52		63

cause of this resonance, the average cross section increases relative to that of a $1/v$ absorber as the average neutron energy increases. This behavior may cause a positive reactivity change with increased temperature. The magnitude of the reactivity change is also a function of temperature. Thus it is possible for the reactor to be stable at low temperatures and to be unstable at elevated temperatures. In order to design the reactor properly, it is necessary to know the exact magnitude of these changes which are quite sensitive to the thermal-neutron spectrum.

Although in intermediate and fast assemblies the thermal-neutron spectrum does not appreciably affect reactivity, it is possible in some reactors of these types to have large thermal-neutron return to the core from the reflector and a consequent large power peak at the core-reflector interface. This peaking may set the limit on heat-transfer performance. Here again, it is necessary to know the thermal spectrum to predict the peaking, although this is a much more complex problem since the detailed spectrum is desired in the vicinity of two dissimilar regions. Similarly, in strongly heterogeneous assemblies, the thermal spectrum is undoubtedly space dependent, and precise knowledge of the spectrum is desirable to predict physics behavior.

Experimental Measurements of Thermal-neutron Spectra^{12,18}

Difficulties involved in the exact theoretical treatment of neutron thermal spectra have necessitated more exact experimental measure-

ments to guide the theory. The experimental measurements are themselves rather complex in that a considerable amount of analysis must be performed to reduce the measured data to the desired form.

For external measurements crystal spectrometers, slow choppers, and pulsed neutron techniques have been used. These external differential measurements have been performed on beams derived directly from the assembly or possibly from a subcritical section of the assembly.

The difficulty involved in these measurements is to obtain the characteristic flux of the assembly. The spectrum must be only a function of the assembly composition and independent of the means chosen to measure it. However, there are a number of ways in which the measuring device may seriously perturb the measured flux. For internal measurements the flux is sensitive to the foil thickness. For the external measurements the flux will be quite sensitive to the size and position of the collimating hole. The energy sensitivity of the neutron detectors used is also an important consideration.

The problem may be illustrated in terms of the pulsed neutron technique,¹⁷ which is a relatively new method but which has furnished the most extensive data for light water highly absorbing homogeneous and heterogeneous assemblies. This method consists of pulsing a neutron source inside a subcritical or purely absorbing assembly. The spectrum at any point in the assembly can be measured by inserting a channel into the point and taking a beam of neutrons from that point. This beam is then analyzed by a suitably arranged time of flight measurement.

For such a measurement to be possible and meaningful, the following are some of the conditions that must be satisfied:¹⁸

1. The slowing-down time of the fast neutrons in the pulse must be much less than the time of flight of the neutrons. This condition ensures adequate energy resolution.

2. The channel must be sufficiently large so that the neutron flux is characteristic of the average assembly composition rather than individual constituents if inhomogeneities are present.

3. The channel must be sufficiently small so that the perturbations of the flux and flux gradients are small.

4. In heterogeneous systems the anisotropic angular distribution around the channel must be known in order to interpret the measured spectra.

5. For small subcritical assemblies the buckling will have to be known. This requirement will usually mean additional measurements.

The same types of complexities are also common to the other methods of measurement.

Measurements of thermal-neutron spectra are furnishing basic information necessary for the understanding of the interaction of thermal neutrons with matter. In addition, the experimental results thus far obtained have established the validity of a gaseous moderator model at thermal energies for light water, since predictions based on this model agree very well with measurements on a $1/v$ absorbing assembly.²⁹ Experiments in heterogeneous lattices have indicated a difference between the spectrum in fuel and moderator which has not yet been attacked theoretically.¹⁷

Theory of Thermal-neutron Thermalization^{18,29}

Theoretical calculations have been limited to those in which the spectrum at a point is due to slowing down of neutrons. Only systems that can be considered homogeneous (heterogeneities small compared to thermal diffusion length) have been analyzed. The solution for thermal-neutron fluxes has primarily concerned itself with the time and space independent problem. For this case, on the basis of neutron conservation, one can write the following integral equation for the flux, $\phi(E)$,

$$\Sigma_a \phi(E) + \phi(E) \int_0^{E_m} \Sigma_s(E \rightarrow E') dE' = \int_0^{E_m} \phi(E') \Sigma_s(E' \rightarrow E) dE' + S(E) \quad (1)$$

where the two terms on the left-hand side of the equation represent the loss of neutrons of energy E by absorption and energy change due to scattering. The first term on the right-hand side of the equation is the gain of neutrons of energy E due to scattering from other energies. The source term, $S(E)$, represents neutrons slowing down from the slowing-down energy region into the thermal energy region. E_m is a cutoff energy defined such that a neutron below the energy E_m has zero probability of attaining energies E_m or greater in a collision with the moderator. For neutrons of energy above E_m , the energy transfer is calculated assuming the moderating atoms to be free from chemical bonds.

The quantity $\Sigma_s(E \rightarrow E')$ is the scattering kernel which, in general, depends on mass of the moderator atoms, temperature, and chemical binding. The kernel is always required to satisfy the condition of detailed balance such that

$$\phi_M(E) \Sigma_s(E \rightarrow E') = \phi_M(E') \Sigma_s(E' \rightarrow E) \quad (2)$$

where $\phi_M(E)$ is the Maxwellian flux distribution. This latter condition guarantees that in the limit, as the source and absorption terms go to zero, the neutrons will reach thermal equilibrium with the moderator and have a Maxwellian energy distribution.

The problem resolves itself into evaluating a scattering kernel $\Sigma_s(E \rightarrow E')$ which will adequately describe the physical situation and satisfy Eq. 2, substituting this value into Eq. 1 and solving for the flux. Although this process is simple to describe, it is not at all easy to carry out. In fact, it has only been done by means of numerical methods for very special models.

In the case of moderation by light and heavy water, the moderating nuclei are bound in a molecule, and the molecules are in turn bound by intermolecular forces. The chemical binding energies are small enough that above a neutron energy of ~ 2 eV each atom can be considered to be free and the interaction can be assumed to be the same as with a monatomic gas. Below this energy binding effects are prevalent, and a collision is no longer an encounter with an individual atom but rather an encounter with an entire molecule, and possibly with a group of molecules at sufficiently low energies. As a result of the molecular binding, the effective

mass of the molecule and, consequently, the energy loss per collision as well as the scattering cross section vary with the energy of the colliding neutron in the low-energy range. It is also possible to have important inelastic scattering by exciting quantum-mechanical vibrational and rotational states in the molecule and liquid. An exact calculation of such a system is of prohibitive complexity. The same is true for scattering by crystalline moderators. Thus it is necessary to use approximate methods.

A practical method is to choose a model that can be calculated and to compare the results with experiment. The simplest model that can be assumed for a moderator is that of a monatomic gas with a Maxwellian velocity distribution at thermal energies. Such a model completely ignores chemical and molecular binding effects. A scattering kernel for this model can be calculated analytically. Such a kernel is known as a Wigner-Wilkins kernel.¹⁹ For light water a particularly simple kernel exists which permits Eq. 1 to be converted to a differential equation that can be solved by numerical methods. The results for light water are in excellent agreement with experiment.²⁰ It is not clear why the agreement is so good except, for reasons that are not understood as yet, the chemical binding has only a small effect. In fact, calculations of the kernel by approximate methods to include chemical binding yield almost identical answers.^{21,22} The gaseous moderator approach also gives good results for heavy crystalline moderators such as beryllium.²⁸

An alternative numerical approach to the calculation of thermal spectra is the Monte Carlo method. This method consists of calculating the detailed behavior of many individual random neutrons as they lose energy and are eventually absorbed and deriving the average behavior from their statistics. For a gaseous moderator model the same result as that given by the Wigner-Wilkins kernel is achieved for light water. The disadvantage of the method is that it is time-consuming compared to a direct numerical approach. However, it does have the decided advantage of being applicable to heterogeneous systems.

The theoretical analyses show that, in principle, the thermal spectrum will depend on the macroscopic cross section Σ_a , the epithermal slowing-down power of the moderator $\xi\Sigma_s$, the moderator temperature, and the thermal energy transfer properties of the moderator. The de-

tailed calculations have shown, however, that the spectrum is quite insensitive to the thermal energy transfer properties and to a good approximation depends only on the ratio $\Sigma_a/\xi\Sigma_s$ and on the moderator temperature. In the limit as $\Sigma_a/\xi\Sigma_s$ approaches zero, the neutron energy distribution is a Maxwellian energy distribution in thermal equilibrium with the moderator. For finite values of $\Sigma_a/\xi\Sigma_s$, the thermal distribution can be characterized by an effective neutron temperature which exceeds the moderator temperature by an amount proportional to $\Sigma_a/\xi\Sigma_s$. However, as $\Sigma_a/\xi\Sigma_s$ becomes of the order unity, the characteristic spectrum loses its resemblance to thermal equilibrium and the concept of a neutron temperature becomes meaningless.¹⁸

Critical Experiments with Highly Enriched Assemblies

During low power experiments which were made with the Pennsylvania State University Reactor³⁰ (modified swimming pool type), criticality was determined for a number of core configurations, which were obtained by loading fuel elements and graphite reflector elements in various patterns in the core grid. Thermal flux distributions were measured for a number of cases, and the reactivity effects of beam hole flooding were determined.

The zero power experiments which were made for the Army Package Power Reactor³¹ are of interest chiefly because they demonstrate some of the characteristics of water-moderated cores of high absorption cross section. Criticality data were obtained, as well as calibration curves for control rods. Effects of control rods on power distribution were measured, and various aspects of the fine structure of the thermal flux were investigated.

*Resonance Escape Probability**

Two reports have recently been issued^{32,33} which give rather comprehensive treatments of the resonance escape probability in thermal re-

* Resonance escape probabilities were measured for water to metal volume ratios of 1.0, 1.5, 2.0, 3.0, and 4.0, with a fuel rod diameter of 0.387 in. The resonance absorption (1-p) in these lattices varied from 51 to 55 per cent of the resonance absorption in uranium metal lattices of similar geometry.

actors containing fertile materials. Also, measurements have recently been reported³⁴ of the resonance escape probability in thorium metal-2 per cent U²³⁵ light water moderated lattices. In view of these activities in the field of resonance absorption, it has seemed worth while to summarize briefly the status of this branch of reactor physics.

Although the resonance escape probability, p , represents one of the earliest concepts introduced into reactor physics, it continues to be the subject of active research. During the past two years, the reliability of the experimental work reported has been considerably increased because of improved instrumentation as well as better experimental techniques. Theoretical work to apply the knowledge has been thereby stimulated, and methods designed to give higher accuracy than has previously been attained have been developed. However, with all the activity in the field, there is still a surprising lack of information available to the reactor designer. As a matter of fact, there is only one set of lattice experiments reported for UO₂ in which p is measured directly, and that for a low density (7.53 g/cm³) fuel element.³⁵ There are no lattice experiments reported for ThO₂.

To illustrate the importance of p , one might consider a low enrichment uranium core with $p = 0.80$. A possible 4 per cent error in p (which is not at all unreasonable) would affect the reactivity by 5 per cent and the conversion ratio by ~14 per cent. The reactivity uncertainty is so large that it would be extremely unwise to build a power reactor without either using a critical facility to check the reactor design or making allowance for possible error by manufacturing additional fuel elements of different sizes or enrichments. Both these alternatives are obviously quite costly. Furthermore, the uncertainty in conversion ratio affects the fuel lifetime in long burning reactors and as such plays a significant role in fuel economics. In addition, the resonance escape probability plays a significant role in determining temperature and void coefficients and so affects directly the safety and stability of the reactor. This latter consideration is a particularly important one in the case of boiling water reactors.

The resonance escape probability is defined as the ratio of the number of neutrons that reach the 1/v region (by slowing down) to the number that originally enter the resonance region. Both fissions and captures occurring in fuel material

should be accounted for, but in the simple four factor formula epithermal fissions are usually omitted and only resonance absorption is taken into account in determining p . This point will be discussed later. This definition can be simply transcribed by diffusion theory into the following:

$$p = \exp \left[-\frac{N_f V_f}{N_m V_m} \frac{\bar{\phi}_f}{\xi \sigma_s \bar{\phi}_m} \left(\int \sigma_0 \frac{dE}{E} \right)_{\text{eff}} \right] \quad (3)$$

where $N_f V_f / N_m V_m$ = ratio of number of fuel atoms to atoms of moderator

$\xi \sigma_s$ = the microscopic slowing down cross section of the moderator

$\bar{\phi}_f / \bar{\phi}_m$ = ratio of the average resonance neutron flux in the fuel to that in the moderator (also termed the resonance disadvantage factor)

$(\int \sigma_0 dE/E)_{\text{eff}}$ = the effective resonance absorption integral

The resonance integral $(\int \sigma_0 dE/E)_{\text{eff}}$ can be fitted by an empirical expression, sometimes referred to as the Wigner formula, which is the sum of a volume term and a surface term, i.e.,

$$\left(\int \sigma_0 \frac{dE}{E} \right)_{\text{eff}} = A \left(1 + \mu \frac{s}{m} \right) \quad (4)$$

where A and μ are constants that are functions of the fuel composition and s/m is the surface to mass ratio of the fuel element, expressed in square centimeters per gram.

In using Eq. 4 to calculate p for a reactivity determination, a modification is usually introduced in which the $1/v$ absorption of the volume term is subtracted out. The reason for the correction is that in using the four factor formula for uranium cores the assumption is made that all fissions in the fuel occur at thermal energies. It is found experimentally that the ratio of epithermal to thermal fissions for U²³⁵ is, to a close approximation, the same as though the fission cross section varied as $1/v$. Thus the effective η including epithermal fissions and $1/v$ absorptions should be the same over the thermal and epithermal energies as it is at thermal energies. Hence, if the epithermal fissions are not included in the calculation, the $1/v$ contribution of the U²³⁸ should be subtracted out to give the proper reactivity. It is also implied that the resonance capture occurs in U²³⁸ independently of the capture in U²³⁵.

A number of methods, described briefly below, have been used to measure the resonance escape probability.³⁵⁻⁴³

Activation Experiments

A measurement of the ratio of the number of Pu^{239} atoms produced by capture in the resonance region to the number of Pu^{239} atoms produced by capture in the thermal region can be used to determine p . This measurement is accomplished by irradiating two foils of low U^{235} content, one cadmium-covered and one bare, in a split rod of the lattice of interest. The number of Pu^{239} atoms produced in the resonance region will be proportional to the Np^{239} activity (2.33 days) of the cadmium-covered foil, whereas the Pu^{239} production associated with thermal neutrons may be obtained from the cadmium difference of the Np^{239} activities. The quantity $(1-p)$ is equal to the fraction of neutrons that undergo resonance absorption. The product of the thermal utilization, which also must be measured, and p represent the fraction of neutrons captured thermally in the fuel rod; this quantity multiplied by the ratio of the macroscopic thermal absorption cross section of U^{238} in the fuel to the total macroscopic thermal absorption cross section of the fuel gives the fraction of thermal absorptions in U^{238} . Thus a simple relation involving p and measurable quantities can be found.

Danger Coefficient

A danger coefficient measurement is made by utilizing a cadmium-covered void near the center of a large reactor. The reactor is brought up to near critical, and the change in reactivity caused by substituting into the void a rod of the material to be investigated is noted. Using a previously calibrated rod as a standard, p and the resonance integral can be quite readily found.

Critical and Exponential Experiments

Using critical and exponential experiments involving heterogeneous lattices the bucklings may be measured. One can then calculate or measure the other pertinent parameters, such as thermal utilization, fast effect, age, and thermal diffusion length, and allow p to be variable to give the proper criticality. This method is an indirect one which can be in considerable error in the absolute value of p since possible errors in the other parameters accumulate in p .

Cross Sectional Measurements

Another indirect method is to infer resonance integrals from the detailed measurements of the neutron cross sections that are now available. Neutron resonance integrals have been derived in this manner for dilute homogeneous mixtures, and estimates have been made of the volume and surface terms of the resonance integral.

In the theory of resonance absorption,^{32,44-54} a number of problems persist at the present time, particularly for heterogeneous reactors, which tend to reduce confidence in calculated results. Some of these problems involve the following:

1. The necessity to estimate cross-section information above the region for which resonances have been resolved. Captures from this upper region constitute about one-third of total in the case of a typical heterogeneous U^{238} system.³²
2. The application of cross-section information involves questions of competition between resonance scattering and absorption. Also the matter of geometrical self-shielding of individual resonances complicates the Doppler broadening effect.
3. The effect of materials surrounding the rod such as cladding and cooling channels is difficult to estimate.
4. The spatial variation of the resonance neutron flux in the moderator and the self-shielding of adjacent fuel elements should be more closely investigated.
5. Degradation of the neutron energy by heavy atoms in the moderator is usually neglected although it is not clear that it should be.
6. The energy variation of the resonance neutron flux in the moderator is assumed to have a $1/E$ variation although it appears that it should be significantly affected by capture in the rod.
7. There is a basic disagreement of the geometric dependence of the resonance escape among the existing theories. The Russian and U. S. approaches are somewhat different, although the results are quite similar over the experimental range in which they have been compared.⁴⁸
8. The effect of epithermal fissions becomes more important as enrichment is increased; it should be accounted for in reactivity as well as being included in p .

These problems will probably be eliminated with further study. However, at present most of the analytical methods that can be applied to reactor design use the phenomenological approach in which some constants are more or less arbitrarily adjusted to agree with experiment. One of the more recent methods of this kind has been reported by Critoph;^{7,55} it has given good results in D₂O moderated lattices.

The Monte Carlo method, a statistical approach, which is becoming more popular as large digital computers are put into operation, has already eliminated some of the previously stated problems. In this method the histories of individual random neutrons are followed through the energy region of resolved resonances. Potentially the Monte Carlo method should yield most of the answers to present day problems. The major difficulties with the method at present are the limitations caused by the uncertainty of the cross-section input data and implicit and explicit errors introduced in the very complex programming for the digital computers. Also, the cost of performing a problem to achieve results that are statistically significant is quite high.

It is important that investigations of resonance absorption continue to the point that ρ can be accurately calculated and applied unambiguously to reactivity calculations. Methods are needed which will permit the design of fuel elements of complex geometrics, such as hollow rods and rod clusters, so that optimum performance, both nuclear and economic, can be attained. At present the only reliable method is to use the rather primitive formulas which are based on experiment and which treat only simple cases.

Oscillations in the Power Distribution Within a Reactor

It has been recognized for some time that oscillations in the spatial distribution of power are possible within a large, high-flux reactor because of the large xenon poisoning effect in such a reactor and because of the time delay (resulting from the holdup in the iodine precursor of Xe¹³⁵) between production and destruction of xenon by neutron flux. In the last few months a number of theoretical studies of this effect have

been made.⁵⁶⁻⁵⁸ The effect is described qualitatively in the following quotation from reference 56:

Consider a reactor which has been operating at a steady flux level sufficiently long to allow the xenon poison to reach its equilibrium value. If a small increase in flux is then made, the destruction rate of Xe¹³⁵ is increased while there is no immediate change in the production rate due to the delay introduced by the I¹³⁵ parent of Xe¹³⁵. Consequently, the small increase in flux causes a decrease in the amount of xenon poison, a corresponding increase in reactivity, and the reactor flux will continue to increase. Similarly a small decrease in flux results in a decrease in reactivity and the flux will continue to fall.

The periods associated with the rise or fall of flux due to departure from the equilibrium condition are such that it is relatively easy to control the reactor power at a set level by manual or automatic control.

Let us now consider a reactor in which the mean power is kept constant using a control rod. If the position of the control rod is altered, for instance due to the insertion of an absorbing sample at some other position in the reactor, a flux distortion is produced. Although the control rod keeps the mean power level constant, the xenon poison in the reactor will decrease in regions where the flux has been increased and will increase in regions where the flux has been decreased. The effect of these changes is such as to tend to emphasize the initial flux distortion due to the insertion of the sample and the corresponding movement of the control rod. It is possible that such flux distortions are unstable, resulting in continued shifting of the flux distribution in the reactor, even though the mean reactor power is kept constant.

Whenever distortions of the flux distribution in the reactor occur, such as those which might be due to the xenon effect, the leakage of neutrons from the reactor is increased above the value which would hold for the unperturbed power distribution. If the reactivity available in the reactor is insufficient to support the chain reaction with the additional leakage, then the spatial oscillation of power cannot occur. This criterion involves not only the available reactivity but also the size of the reactor and the migration area of neutrons in the reactor since these quantities determine the neutron leakage for a given flux distribution. The situation may be looked at in more mathematical terms in the following way. Ordinarily, the flux distribution in the reactor is described in terms of a set of mathematical functions which are solutions of the wave equation, $\nabla^2 \phi + B^2 \phi = 0$, and which satisfy the boundary conditions at the boundaries

of the reactor. For example, if the reactor has the form of an infinite slab of extrapolated thickness a in the X direction, the flux distribution may be described in terms of the set of functions $\sin n\pi x/a$. If the slab reactor is of uniform composition and is just critical, the flux distribution will be given by $\varphi = \sin \pi x/a$ (the origin is taken at one of the extrapolated boundaries of the slab). As the condition of criticality, the "geometric buckling," $(\pi/a)^2$, must be equal to the "material buckling," which, according to the simple one-group picture, is $k - 1/M^2$, where M^2 is the migration area and k is the infinite multiplication factor. If the nonuniformity in xenon distribution causes a distortion of the flux distribution from this so-called fundamental mode distribution, the new distribution will be described in terms of higher modes (i.e., $n > 1$). Reference 56 shows that in order for an unstable flux oscillation to occur, which involves the j th mode for its description, it is necessary that the unpoisoned reactivity be sufficiently high to make the reactor critical in the j th mode. That is to say

$$k - 1 > B_j^2 M^2$$

where B_j^2 is the geometric buckling for the j th mode. In the case of the slab reactor, for example, the second mode is that for which $n = 2$, or $B_2^2 = (2\pi/a)^2$ and the unstable oscillation would occur only if $(k - 1)_{\text{unpoisoned}} > (2\pi/a)^2 M^2$. This condition is a necessary one but not a sufficient one except for reactors of sufficiently high thermal neutron flux that the destruction of xenon by neutron absorption is much faster than the destruction by radioactive decay.

Reference 56 estimates that the NRU reactor, although stable in the higher modes, in the sense that perturbations are damped out, can be subject to transient flux distortions which can be appreciably larger than the initial flux distortions caused, e.g., by movements of control rods. It also states that if the reactor were held down in its most reactive condition by control rods, all of which were installed on a diametrical plane, violent oscillations of power between the two halves of the reactor could occur.

Reference 57 analyzes the problem by a different technique and investigates also the effect of a negative temperature coefficient of reactivity in stabilizing the flux distribution. It suggests also that, if the possibility of flux oscillations is recognized in a given reactor during the design period, provision may be made for

controlling the tendency to oscillate by judicious placement of control rods. It also points out that such an oscillation could be a design limitation in the case of reactors designed to use some form of control such as soluble poison, whose position in the reactor cannot be adjusted.

Briefly Noted Reports

Reference 59 is a study of the Pu^{240} capture cross section, treating experimental evidence of various types. The important effects of self-shadowing of samples used for measurements is pointed out.

Reference 60 is a review of the experimental information on α of Pu^{239} . A "presently suggested" form of this dependence is obtained which is shown to be consistent with nearly all measurements.

Reference 61 reports experimental measurements of abundances and half lives of delayed neutrons from the fast fission of U^{235} , U^{233} , U^{238} , Pu^{239} , Pu^{240} , and Th^{232} . Godiva was used as the neutron source.

Reference 62 reports calculations of fission-product spectra in discharged fuel from fast-fission power reactors. Reactor operating times from 10 to 10,000 days and cooling times from zero to infinity are covered.

Reference 63 contains computed tables of Xe^{135} concentration build-up in reactors operating at various specific power levels.

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Recent SPERT Experiments

Further experimental investigations of the dynamic behavior of the SPERT-I reactor were reported at the third annual ANS meeting. The results of the experiments have not been fully analyzed, and hence the reported results^{1,2} were limited to an account of the actual over-all observations. In most of the experiments reported, the excess reactivity Δk was introduced into the reactor system as a terminated ramp to different values of total reactivity. The ramp was introduced at a rate of about 0.09 per cent $\Delta k/k$ per second. The effect of the ramp addition of reactivity is to cause the reactor power to rise until the rise is terminated by the inherent power limiting characteristic of the reactor. For large total additions of reactivity, the self-limiting mechanism is boiling of the water in the reactor core. For sufficiently large total reactivity additions (about 2.5 per cent or more), the reactor power may not approach a steady value after the initial power surge but may go into an unstable mode of operation characterized by oscillations of rapidly increasing amplitude. This behavior appears to have many characteristics in common with the instabilities observed in boiling reactors when the reactivity compensated by steam becomes too large, and, in fact, it is probably the same phenomenon.

Experiments were conducted to determine the effects, on the oscillatory behavior of the reactor during the transient, of the following variables: (1) change in height of water over core from 2 to 9 ft, (2) blocking off of approximately one-third of the fuel element channels in the core, (3) change in surface tension of the water, and (4) increase in gas content of the water.

In an over-all sense the SPERT-I reactor exhibited the following behavior for ramp additions of reactivity:

1. Increased water head showed a definite increase in tendencies to oscillate in a divergent manner and generally resulted in increased amplitudes of oscillation.

2. Blockage of a portion of the flow channels showed surprisingly little effect. The only im-

portant effect appeared to be a 15 per cent decrease in repetition rate of the oscillations.

3. A 35 per cent reduction in water surface tension through the addition of a detergent had a quieting effect on the oscillations. There was a tendency for more complete damping or smoothing out of the oscillations in the post-initial-burst behavior of the reactor. The integrated energy release was essentially unchanged. However, in the experiments with the detergent, there was appreciable frothing of the water which perhaps masks the true effects of change in surface tension.

4. There was no essential difference in the tests due to change in initial power of the reactor prior to the excursions. The range of initial power investigated was 5 watts to 100 kw; the period range, from 2 sec to 23 msec.

In assessing the effect of the water height above the reactor core, experiments in which reactivity is added effectively instantaneously are perhaps more significant than those in which ramp additions are used, because there is reason to expect that the additional inertia of the water in the submerged case may result in considerably higher transient peak powers. Experiments of this type showed that when the applied excess reactivity was relatively small the peak power was higher by about a factor of two for the deeply submerged core but that as larger values of applied reactivity were used the difference between deep and shallow submergence decreased. When the applied reactivity was sufficiently large to cause a power rise with an exponential period of 9 msec, the difference between the peak powers in the two cases was only 25 per cent. This result is a very encouraging one since it indicates that the results of the BORAX experiments and the earlier SPERT experiments can be applied to submerged reactors such as swimming pool reactors without large errors. A further result of experiments using "instantaneous" reactivity additions was that an increase in the dissolved CO_2 content of the water appeared to have no significant effect on the power transient.

A theory was reported^{3,4} which was developed to fit mathematically the oscillatory behavior

observed in SPERT-I reactor. The theory is based on an assumed void growth equation, involving two adjustable constants, and the usual pile kinetic equations. The choice of the values of the two adjustable constants was made to give the observed period for a single observed SPERT oscillation; the constants are not derivable from the thermal-hydrodynamic processes actually involved. With the chosen constants, the theory agrees fairly well with the over-all behavior of the SPERT-I transients observed in the tests.

Analysis of the Final

BORAX-I Experiment

Final destructive experiment with the BORAX-I reactor has been examined⁵ in the light of recent experimental data on aluminum-water explosions. Experiments indicate that it is necessary to have the aluminum dispersed in the water and at a temperature of at least 1600°C in order to produce a high intensity metal-water explosion. It is concluded from the study of the BORAX-I observations that this temperature was evidently not reached. It is evident, however, that the temperature could not have been greatly less than 1600°C, and hence the further conclusion is drawn that a slightly greater release of nuclear energy might have produced a very substantial chemical energy release.

Blast Effect Studies

At the third annual ANS meeting results of experimental blast effect studies on a one-quarter scale model of the Air Force Nuclear Engineering Test Reactor were presented.⁶ In the tests charges located at the reactor core position were fired, and the pressure, strain, and displacement histories were measured at various locations. The charge strengths ranged from 26 to finally 1000 Mw-sec equivalent energy release. Both explosives and propellants were used as charges to give different rates of energy release, although in both cases the rates are stated to be much higher than those indicated in the BORAX destructive test. The overall results of the tests are briefly outlined as follows:

1. At 202 Mw-sec energy release hairline cracks developed in the concrete shield.

2. At 278 Mw-sec the steel beam ports were distorted but remained intact.

3. At 444 Mw-sec the 10-12 bolts holding down the reactor vessel top plug were fractured.

4. At 1000 Mw-sec the same bolts fractured and were projected at a velocity of 130 ft/sec. The welds in the irradiation cells were cracked. The bridge over the shielding test pool adjacent to the reactor was thrown up into the air. The outer containment wall of the building was breached.

The experiments suggest certain modifications in the reactor containment structure. With these modifications, the authors estimate that the reactor containment structure will safely contain an explosively released energy of 1000 Mw-sec.

These experiments are of particular significance because they set a precedent for the large-scale destructive testing of reactor containment. If experiments of this type can lead to rational and reliable methods for designing containment structures, they may contribute a great deal to the advancement of the nuclear energy technology. However, if it becomes the practice to conduct such tests for new reactor installations, it is important that the reliability and applicability of the tests be well established. Otherwise they may represent an important increase in the already large cost of reactor safeguard without yielding a compensating improvement in confidence of containment.

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General Discussion

Most of the published data on fuel elements are concerned with the manufacture and testing of fuel elements which will have a high degree of corrosion resistance in the coolant media and good radiation stability at high burn-ups. In this regard there is a strong temptation to incorporate materials of relatively high absorption cross section, which may make for poor neutron economy in the reactor.

The first issuance of *Power Reactor Technology*¹ contains a general discussion of fuel element characteristics as applied to large central station power reactors. A summary of this discussion as a guide in evaluating the fuel elements is included below.

Relation of Fuel Element to Fuel Cost

1. *Use Charge.* This is a capital charge against the value of fuel in the inventory necessary for reactor operation. Factors which favor low use charges are: inexpensive fuel (e.g., a U^{235} - U^{238} mixture of low enrichment); high specific power; low inventory in fabrication and reprocessing; and high net thermal efficiency.

2. *Fuel Burn-up Charge.* This is an operating expense equal to the net value of fuel destroyed per kilowatt hour of electricity produced. The factors favoring low burn-up cost are: use of cheap fuel; high conversion ratio; long fuel lifetime; and high net thermal efficiency.

3. *Fuel Fabrication Cost.* This is a capital charge against fuel inventory and an operating charge for fuel element replacement. Fuel fabrication costs can be minimized by optimizing the design of the fuel element for long fuel lifetime (with respect to fission damage, corrosion, and reactivity), low fabrication cost, and maximum neutron economy.

Fuel Element Lifetime

For most irradiation tests on fuel elements, fuel lifetime is expressed in percentage burn-up

of the fuel atoms. For a mixture of U^{235} - U^{238} metal, the relation among various units of lifetime is:

1 per cent burn-up of fuel

~ 0.84 per cent of fuel atoms fissioned

~ 0.01/E atoms "burned" per initial fissionable atom

~ 0.0084/E atoms fissioned per initial fissionable atom

~ 6950 Mwd/ton

where E = initial enrichment = atoms of fissionable isotope/total fuel atoms, and the useful energy release per fission has been taken as 190 mev.

Absorption by Additives

Although the ideal fuel element from the nuclear physics viewpoint should contain only fuel (fissionable plus fertile material), additives are often necessary for improvement of corrosion performance, irradiation lifetime, and other characteristics. For maximum neutron economy the absorption by additives must be kept very low, even when enriched fuel is used. Good neutron economy favors low fuel cost.

Fuel Density in the Element

The most important effect of density is the increase in surface to mass ratio with decrease in density for a given fuel element size. This increase causes a higher relative parasitic absorption in fuel cladding and process tubes. It also requires, for a given resonance escape probability, that a lower mass ratio of fuel to moderator be used and thus results in higher relative absorption by the moderator. Finally, it results in a lower fast-fission effect.

Uranium Metal and Alloys of High Uranium Content

Recent development and testing of elements of this type are summarized below:

Natural Uranium

A single representative curve (Fig. 3) for thermal conductivity of ordinary purity beta treated uranium has been prepared by Argonne National Laboratory (ANL).^{2a} The curve is a composite of what appears to be the best available values to date in the temperature range from absolute zero to 900°C. The author, however, cites that more information and confirmation of data on this subject to eliminate serious deficiencies and uncertainties is required.

A second part of this report, issued separately,^{2b} presents data on the electric conductivity of uranium, including the British work on purity, a study of transition temperature effects, and other more unusual points, like the effects of annealing temperature, cycling growth, preferred orientation, and hydrostatic pressure.

Room-temperature tensile properties for a number of specimens of irradiated natural uranium were determined by Hanford Atomic Products Operation (HAPO).³ Specimens were exposed to burn-ups as high as 0.10 at. % (total) at ~150°C. The properties of the longest exposure specimens were nearly the same as the short exposure specimens, indicating that the damage is essentially saturated at low exposure levels. Table III-1, reproduced from the reference report, summarizes the effect of irradiation and post-irradiation annealing on the room-temperature tensile properties of uranium.

Depleted Uranium

Depleted uranium (0.028 wt. % U^{235}) specimens were irradiated to 0.32×10^{20} and 3.35×10^{20} nvt, corresponding to burn-ups of ~0.00052 and ~0.0048 per cent, or 1.3×10^{18} and 1.2×10^{19} nvt integrated neutron flux in natural uranium.⁴ The lower exposure specimens were straight and smooth, and the higher exposure specimens were warped and roughened.

Uranium and Uranium Alloys Prepared by the Powder Metallurgy Technique

In the studies of dimensional stability of uranium metal under irradiation, it has been found that the irradiation stability is greatly influenced by the orientation and size of the grains in the material. To eliminate certain of these disadvantages, powder metallurgy techniques

have been applied to the manufacture of uranium and uranium alloys. A study⁵ of the effects of irradiation on powder compacts of uranium and uranium alloys has recently been completed by ANL. The results of this study, details of which can be found in the reference report, are summarized in the report as follows.

Irradiation tests were made on powder compacts prepared from uranium, uranium hydride, and the following alloys: uranium-1.0, 1.4, 2.0, 2.5, and 3.5 wt. % molybdenum; uranium-1.6 wt. % niobium; and uranium-0.42 wt. % silicon. The compacts were prepared by end-pressing, side-pressing, hot-rolling, and hot-swaging. When subjected to irradiation, the uranium compacts invariably shortened in the pressing direction; the compacts prepared from uranium

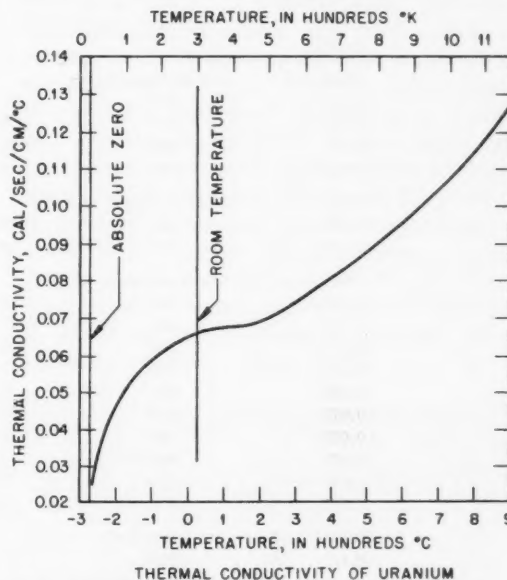


Figure 3—Representative curve for ordinary purity beta-treated uranium along rolling direction.

hydride elongated in the pressing direction; and the alloy compacts elongated in the direction of pressing, rolling, or swaging. The surface smoothness of the irradiated molybdenum alloy compacts improved with increasing molybdenum content, although dimensional stability tended to decrease. The irradiated niobium alloy showed comparatively good surface smoothness and dimensional stability, but the silicon alloy specimens were badly roughened and elongated. Both the unalloyed and the alloy compacts decreased in density under ir-

radiation, and the rate of density decrease was found to depend strongly on the irradiation temperature.

Uranium-Titanium*

The following conclusion has been made as a result of an investigation^{6a} to improve the mechanical properties and corrosion resistance of uranium by small additions of titanium:

1. Small additions of titanium to uranium im-

3. The mechanical properties of alloys of uranium containing small additions of titanium are very dependent on the rate of cooling applied after a heat-treatment in the gamma region of temperature.

4. Optimum mechanical properties—tensile and yield strength, ductility, and impact strength—are obtained from additions of 0.5 to 1.0 wt. % titanium in gamma-water-quenched condition.

5. The solid solubility of alpha uranium for

Table III-1 EFFECT OF IRRADIATION AND POST-IRRADIATION ANNEALING ON THE ROOM-TEMPERATURE TENSILE PROPERTIES OF URANIUM*

R. S. Kemper and W. S. Kelly

Exposure, at. % burn-up	Annealing temp., °C	Annealing time, hr	Ultimate strength, 1000 psi	0.2% yield strength, 1000 psi	Elongation, % in 1 in.
Control			97.1	37.8	19 ± 1
0.018	As irradiated		78.0†	57.0†	0.75†
0.018	400	10	75.0	39.2	2.5
0.018	400	25	76.0	39.2	2.6
0.018	400	100	74.9	48.1	1.6
0.018	600	10	84.5	35.2	5.7
0.031	As irradiated		83.0†	73.0†	0.7†
0.031	400	100	79.2	56.6	1.1
0.031	600	10	82.3	42.2	2.7
0.075	As irradiated		88.0†	79.0†	0.5†
0.075	400	10	80.7	62.8	0.7
0.075	400	25	77.7	57.2	0.7
0.075	400	100	82.3	63.5	0.8
0.075	600	10	87.5	53.0	1.2
0.075	600	25	81.0	70.5	0.9
0.075	600	100	87.2	50.6	1.2
0.075	700	10	52.5†	40.8†	0.95†
0.075	700	3†	40.0	24.3	1.14
0.075	700	10‡	49.1	18.7	2.1
0.075	800	10‡	36.1	17.3	12.4
0.10	As irradiated		77.4†	73.4†	0.55†
0.10	800	10‡	32.2	29.1	4.6

* Data taken from Report WASH-703.

† Average.

‡ Heated at 10°C/hr held 3 hr, furnace cooled (~150°C/hr).

§ Heated and cooled from 600°C/hr.

prove its mechanical properties and resistance to corrosion.

2. The range in which useful improvement is obtained lies between 0.1 and 1.5 wt. % titanium addition. Below this range, properties are essentially those of unalloyed uranium; above it, high strength is accompanied by brittleness, low ductility, and low impact strength.

* Parasitic absorption cross section: $\bar{\Sigma}_p = (0.0115 \text{ cm}^{-1})(\text{wt. \% titanium})$.

titanium is in the range of 0.5 to 1.0 at. % rather than 4 at. % as reported in recent constitutional diagrams.

6. The extent of the beta-uranium phase field is less than 2.5 at. % titanium rather than 4 at. % as reported in recent constitutional diagrams.

7. Precipitation hardening treatments improve tensile and yield strengths of uranium containing small additions of titanium but de-

crease sharply its ductility and impact strength.

A study⁷ conducted by ANL indicated that 2.7 wt. % titanium in uranium is the minimum required for reasonable corrosion rates at 290°C. A summary of the corrosion behavior of the gamma-quenched uranium-2.7 wt. % titanium alloy at 290 and 315°C is included in Table III-2. Previous data on uranium-titanium alloys have been reported in reference 6a.

Table III-2 CORROSION OF GAMMA-QUENCHED URANIUM-2.7 WT. % TITANIUM ALLOY*

Temp., °C	Time, days	Corrosion rate, Mcd (period)	Comments
290	5.0	24.0	Uniform corrosion
	5.8	31.8	
	5.9	39.0	Sample appears to be dividing into layers
	6.9	54.4	
315	3.8	77.2	Pitting
	6.8	116.2	Test stopped

* Data taken from Report ANL-5643.

Uranium-Silicon

A fuel material of interest is the intermetallic compound U_3Si which contains 3.8 wt. % silicon. Several specimens of this compound were irradiated in NaK capsules in Materials Testing Reactor (MTR) to burn-ups listed in Table III-3, and two specimens were checked for corrosion after irradiation.^{6b} The castings were very stable, the length changes being between 1 and 2 per cent for all three specimens which were 1 in. long. The corrosion resistance of the specimens was relatively unaffected by the irradiation.

Uranium-Molybdenum Alloy

Recent data on experiments by Atomics International on uranium base fuels for the Sodium Reactor Experiment (SRE) show that the radiation dimensional stability of powder compacted uranium-1.2 wt. % molybdenum is relatively poor.³ A group of specimens were irradiated at the MTR in NaK capsules at maximum central fuel temperature of 750 to 1200°F and to burn-ups of 0.4 at. % (total max.). Diameter increases of ~8 to 12 per cent were noted. A summary^{3,8} of these data is compiled in Table

III-4. Of the eleven specimens tested (eight included in the table), three of them showed uranium-iron reactions and could not be disassembled. The reaction occurred between the fuel alloy and an internal steel sleeve.

It has been reported previously that the uranium-10 wt. % molybdenum alloy was found to have the best radiation stability of the alloys (uranium-5, 7, and 10 wt. % molybdenum) tested.¹

Uranium-Zirconium-Niobium

A study⁹ of the high-temperature aqueous corrosion of uranium alloys containing minor amounts of niobium and zirconium has recently been completed by ANL. The uranium-5 wt. % zirconium-1.5 wt. % niobium alloy, quenched from the gamma phase, has good corrosion resistance in degassed, distilled water up to temperatures of 315°C. However, the corrosion resistance is destroyed by over-aging at temperatures as low as 350°C. Experimental evidence indicates that corrosion failure is due to absorption of hydrogen. The above alloy can be made fairly dimensionally stable under irradiation by heat-treatment, but the corrosion resistance is destroyed in the process.^{1,10} Irradiation tests¹⁰ also show that the corrosion resistance of this alloy is destroyed after a burn-up of between 0.046 and 0.074 at. %.

Table III-3 EFFECT OF IRRADIATION ON U-Si ALLOYS

Specimen	Burn-up U atom	Mwd/ton	Irrad. temp., °C	Corrosion time, hr	Corrosion rate, mg/cm ² /day
1	0.0	0	0	137	5.3
2	0.12	1000	280	86	0.72
3	0.33	2800	630		
4		0	0	88	16.0
5	0.71	6000	860	87	32.0

Uranium-Niobium

A study¹¹ of the effect of impurities in uranium-10 wt. % niobium alloy (not completely homogeneous) on its corrosion behavior in 680°F degassed high purity water indicates:

1. Corrosion resistance of gamma-phase base alloy samples was: (1) decreased by increasing additions of carbon from 190 to 1700 ppm and (2) not significantly affected by additions of 90 ppm beryllium, 40 ppm hydrogen, 270 ppm ni-

trogen, 720 ppm oxygen, 700 ppm tantalum, 3500 ppm tin, and 850 ppm titanium.

2. Corrosion life of Zircaloy-2 clad samples containing an intentional defect was much longer than that of the bare alloy. Some specimens survived more than 300 days exposure. The

Table III-4 RADIATION EFFECTS ON
URANIUM ALLOY FUEL^{3,9}

(Nominal sample size, 0.375 in. diameter by 1.500 in. long)

Composition of specimen	Max. measured central temp., °F	Estimated total burn-up, at. %	Av. diam. increase, %
U-1.2 wt. % Mo*	1250	0.2	8.0
U-1.2 wt. % Mo*	1110	0.2	11.0
U-1.2 wt. % Mo*	970	0.4	12
U-1.2 wt. % Mo*	750	0.3	9
U-2.0 wt. % Zr†	600	0.25	4
U-2.0 wt. % Zr†	910	0.4	§
Unalloyed U‡	960	0.4	§
Unalloyed U‡	900	0.4	§

* Powder compacted.

† Cast.

‡ α -rolled, BHT.

§ Specimen could not be removed from capsule for measurement.

corrosion resistance of clad samples was decreased by impurity additions in excess of 800 ppm of carbon, oxygen, tantalum, tin, or titanium.

3. Combinations of carbon, nitrogen, and oxygen had little more effect on corrosion behavior than each of the impurities above.

Uranium-Zirconium

In conjunction with the fast breeder reactor program, ANL has made irradiation tests on fuel pins using uranium-2 wt. % zirconium alloy.^{6b,6c} The pertinent data for these irradiations are tabulated in Tables III-5 and III-6.

The results indicate 2 per cent Zr-U alloy fuel pins, as cast, are stable to 0.6 at. % fuel alloy burn-up, provided the alpha-beta transformation temperature (663°C) of unalloyed uranium is not exceeded. Even occasional cycling above this temperature causes instability, re-

flected in pronounced bumping of the fuel and its container.

Specimens irradiated above 600°C to burn-ups of over 1 per cent, as shown in Table III-7, undergo severe swelling of the specimen diameter.^{6b}

The swelling is believed to result from the accumulation of fission gases and the formation of a large central void. An axial hole placed in one of the specimens (No. 3) to relieve this condition resulted in very little swelling, even though the irradiation temperature was 350°C higher than the solid specimens of Nos. 1 and 2.

Uranium-Fissium Alloy

Irradiation tests have been conducted on reference design fuel elements for a sodium cooled, fast breeder reactor (EBR-II) (reference 6c). The fuel, 0.145-in.-diameter pins by 14.22 in. long, is canned in 0.008-in. wall 304 stainless-steel tubing. A minimum growth gap of 29 per cent is provided, and a 0.006-in. annulus between the fuel and tubing is filled with sodium for heat transfer. The first loading for EBR-II will use a uranium alloy and not a uranium-plutonium alloy fuel element. A summary of pertinent data on the tests appears in Tables III-5 and III-6.

The fissium alloys of uranium when cycled through the alpha-beta transition temperature of unalloyed uranium are very stable compared to the 2 per cent Zr-U alloys which showed severe instability under this irradiation procedure.

Uranium-Plutonium Alloys

The uranium-plutonium alloys are of primary interest in the design of fast breeder reactors. Tentatively, the specification for the fuel for such a reactor (EBR-II) calls for the fuel to be dimensionally stable to burn-ups of at least 2 wt. %, or 20,000 Mwd/ton, at operating temperatures of 700°C. In addition, the fuel should contain at least 20 per cent plutonium for optimum breeding gain.

Specimens of uranium-5, 10, 15, and 20 wt. % plutonium alloy specimens have been irradiated to high burn-up.^{1,6b} The best irradiation stability was found with uranium-20 wt. % plutonium and uranium-20 wt. % plutonium-5 wt. % fissium. (Fissium, Fs, composition is nomi-

nally 4.6 wt. % zirconium, 25.9 wt. % molybdenum, 39.8 wt. % ruthenium, 6.5 wt. % rhodium, and 23.2 wt. % palladium.) Table III-8 is a summary of the irradiation data.

reported. Table III-9, extracted from the reference report, gives some of the details of the irradiation tests and their effect on the dimensional stability of the specimens.

Table III-5 EBR-II FUEL ELEMENT IRRADIATIONS

Test No.	1	2	3	4	5	6	7	8
Fuel geometry*	7 slugs		1 pin		6 slugs	1 pin		3 slugs
Fuel length, in.	13.4				14.22			
Fuel diameter, in.	0.164				0.144			
Alloy composition†	2.0 Zr-U 1.3 Zr-U			2.0 Zr-U			Fissium 4	Fissium 3
Fabrication‡	As cast	Wrought and gamma treated			As cast	Wrought and gamma treated		
Enrichment, %	15	17	20	17			20	
Bond thickness, in.	0.004					0.006		
Gas reservoir, in.§	2.5			None			1.5	2.5
Growth gap (% of total length)	3.2				2.0			

* Slugs indicate more than one piece used to obtain fuel length.

† Fissium 4: 96% U, 2.0% Mo, 1.2% Ru, 0.4% Pd, 0.25% Rh, 0.15% Zr.

Fissium 3: 96.9% U, 1.55% Mo, 1.15% Ru, 0.1% Pd, 0.2% Rh, 0.1% Zr.

‡ Tests 1 and 5: Statically cast, centerless ground to size (~5% reduction).

Tests 2, 3, 4, and 6: Rolled, swaged to size, heated to 800°C, furnace cooled.

Tests 7 and 8: Statically cast, swaged to size (~5% reduction) at 800°C, air cooled.

§ Tests 1, 2, 6, 7, and 8: An expansion space is provided within the element.

Tests 3, 4, and 5: Element open at the top, filled with sodium, expansion is into the finned capsule.

Table III-6 EBR-II FUEL ELEMENT TEST CONDITIONS

Test element No.	Operating fuel temp., °F	No. of thermal cycles	Estimated burn-up, at. %
1	1000-1050	140	0.4
2	775-825	210	0.45
3	725-775	260	0.75
4	825-875	290	0.7
5	625-675	220	0.6
6	625-675	200	0.6
7	625-675	>250	1.0
8	625-675	>250	1.0

The tests on the cast U-Pu elements containing up to 15 wt. % plutonium were unsatisfactory as shown in the table for specimens 1, 2, and 3. Extrusions of these alloys showed considerable improvement over the castings with specimen 6, containing 20 wt. % plutonium, showing good dimensional stability.

ANL has recently reported the results of an irradiation study of cast uranium-plutonium base alloys.¹² Good dimensional stability and surface finish of the specimens tested were

Table III-7 EFFECT OF IRRADIATION ON URANIUM-2 WT. % ZIRCONIUM

Specimen No.	Zr, wt. %	Total burn-up, at. %	Irradiation temp., °C	Remarks
1	2	6.6	660	Considerable swelling
2	2	2.8	700	Considerable swelling
3	2	2.0	1020	Very little swelling

Thermal expansion studies⁷ by ANL of uranium-plutonium-fissium alloys have revealed an erratic behavior when heated above 500°C of fissium alloys having a molybdenum content of 5 wt. %. This alloy (uranium-20 wt. % plutonium-4.98 wt. % molybdenum-0.25 wt. % zirconium-0.35 wt. % rhodium-1.25 wt. % palladium-2.15 wt. % ruthenium), when cycled to a maximum temperature of 713°C, had a phase change which started at 541°C. A shrinkage occurred in the specimen amounting to a total decrease in length, at room temperature, of about 0.003 in. in 0.5 in. In subsequent cycles

the phase change settled in the temperature region of 527 to 548°C. After the initial shrinkage the specimen progressively grew on each cycle until on the eleventh cycle it was longer than the original specimen length.

Table III-8 EFFECT OF IRRADIATION ON Pu-U ALLOYS

Specimen No.	Pu, wt. %	Total burn-up, at. %	Remarks
Cast			
1	5	0.8	Severe surface roughening
2	10	1.2	Severe surface roughening
3	15	1.7	Severe surface roughening
Extruded			
4	10	0.70	Dimensional changes
5	15	0.32	Dimensional changes
6	20	0.40	Good surface and dimensional stability

Thorium and

Thorium-Uranium Alloys

Recent data⁷ on irradiation tests on thorium and thorium-uranium alloys have been reported by ANL. A group of specimens were irradiated in NaK capsules in the MTR for 32 months to burn-up as high as 4.4 at. %. The dimensional stability and surface smoothness of the specimens remained excellent. Table III-10 is a summary of the irradiation effects on these fuel materials.

Nonmetallic Elements of

High Fuel Content

Urania (UO₂)

Significant properties of UO₂, along with detailed information on corrosion and radiation stability, were covered in the first issue of the journal.¹ The quantity of fission gas released in UO₂ fuel under irradiation is particularly important. Results of tests previously reported indicate that with two specimens, one 91 per

cent and the other 95 per cent of theoretical density (10.96 g/cm³), the fission gas released after irradiation to 3000 and 5000 Mwd/ton at central temperatures of ~1600°C was ~3.1 per cent and 1 per cent for the two specimens.^{13,14}

A study on the mechanism of fission gas release from UO₂ has been made by Bettis Plant.¹⁵

The general conclusions and results of the investigation indicate:

1. The release of fission gases from irradiated UO₂ is in all probability controlled by diffusion of the gases from the oxide matrix.

2. The oxide density has a large effect on the gas release because of the variation with density of the diffusion path length to the external surfaces. (Experimental evidence indicates that the fission gas release is reduced considerably with UO₂ specimens which are >90 per cent of theoretical density.)

3. Assuming the gas release is due to diffusion, the amounts of gas released can be calculated with a correction being made for the density of the oxide specimen. Calculations agree within a factor of 3 with experimental data on irradiated Pressurized Water Reactor (PWR) fuel rods.

UO₂-SiC-Si

This ceramic fuel element is being studied because of its potential use as a high-temperature reactor fuel. Table III-11 shows the results of some irradiation studies made by Oak Ridge National Laboratory (ORNL).¹⁶

The specimens showed good dimensional stability, and photomicrographs revealed no cracking. Tests were made to determine clad efficiency. Fuel plates were irradiated at elevated temperatures, and the resultant fission gases in the irradiation capsule were analyzed. Xe¹³³ was the only significant gas detected, and it represented only 10⁻³ per cent of the total Xe¹³³ being produced in the sample at the end of the irradiation period.

Thoria-Urania (ThO₂-UO₂)

ThO₂-UO₂ ceramic fuel specimens are being studied for use in high-temperature reactors as a thermal breeder fuel material. Some recent irradiation history on this fuel material reported by ORNL is shown in Table III-12 (reference 16).

Table III-9 SPECIMEN DATA ON URANIUM-PLUTONIUM BASE ALLOYS*

Specimen No.	Composition, wt. %	Approx. calc. max. temp., °C	Calc. burn-up of all atoms, %	Changes due to irradiation					
				Length increase, %	Diam. increase, %	Calc. volume increase, %	Hardness change, R _A	Weight change, mg	Density decrease, %
CF-1	U-20 Pu-5 Mo	340	0.27	0.74	0	0.74	-8.3	-4	0.74
CF-2	U-20 Pu-5 Mo	280	0.43	0.67	0.56	1.80	-8.3	-16	1.45
CF-3	U-20 Pu-5 Mo	270	0.20	0.79	0.48	1.76	-8.3	2	0.68
CF-4	U-20 Pu-5 Mo	260	0.38	1.26	0.40	2.07	†	1	0.62
CF-5	U-20 Pu-5 Mo	190	0.28	0.83	0.48	1.80	-23.9	1	1.08
CF-6	U-20 Pu-5 Mo	120	0.07	‡	0	†	-17.2	†	†
CF-7	U-20 Pu-5 Mo	270	0.40	1.05	0.96	3.00	-7.0	0	1.59
CF-8	U-20 Pu-5 Mo	220	0.15	0.77	0.16	0.40	-7.7	0	0.23
CF-10	U-20 Pu-10.8 Fs	310	0.92	2.30	0.16	2.63	†	-1	1.29
CF-12	U-20 Pu-10.8 Fs	280	0.81	0.88	0.40	1.69	†	-1	0.18
CF-14	U-20 Pu-5.4 Fs	560	0.42	‡	1.80	†	†	†	0.46
CF-15	U-20 Pu-5.4 Fs	520	0.40	0.57	0.89	2.37	†	-1	0.91
CF-16	U-20 Pu-5.4 Fs	450	0.33	0.62	0.08	0.78	†	-3	0.28

* Data taken from Report ANL-5677.

† Breakage by indenter.

‡ Loss of material.

Table III-10 EFFECT OF IRRADIATION ON THORIUM AND THORIUM-URANIUM ALLOYS*

Specimen No.	Method of preparation	Uranium,† wt. %	Indicated burn-up, wt. %	Calc. max. temp., °C		Length change, %	G ₁ , microin./in./ppm burn-up	Diam. change, %	Density change, %	Hardness change, R _A
				Surface	Center					
H-10	As swaged at 25°C	0	2.3	76	88	-0.087	-0.038	1.42	-0.612	16.9
H-14	As swaged at 25°C	0	2.7	82	96	+0.18	+0.067	0.68	-1.09	16.3
H-15	As swaged at 25°C	0	2.9	86	101	+0.15	+0.052	0.63	-1.45	16.0
H-5	Swaged at 25°C, 2 hr at 700°C	0.13	1.7	45	49	+0.47	+0.27	0.63	-1.07	26.2
H-4	Swaged at 25°C, 2 hr at 700°C	0.13	1.9	55	63	+0.53	+0.28	0.84	-1.40	27.2
H-1	Swaged at 25°C, 2 hr at 700°C	1.39	2.4	90	104	+0.67	+0.28	1.11	-1.45	7.9
H-8	Swaged at 600°C, 2 hr at 700°C	5.53	2.8	108	130	+0.17	+0.060	1.74	-3.56	15.9
H-7	Swaged at 600°C, 2 hr at 700°C	5.53	4.4	160	200	+0.27	+0.061	2.58	-4.79	15.0

* Data taken from Report ANL-5643.

† 93% enriched. The balance of wt. % in column is thorium.

UO₂-Graphite

Studies have been conducted at Battelle Memorial Institute (BMI) to determine uranium losses and fission-product retention of UO₂-graphite elements in a flowing nitrogen atmosphere.¹⁷ Data on the uranium losses are shown in Table III-13. Results on the fission-product retention will be reported at a later date.

Table III-11 SiC-UO₂ SPECIMEN HISTORY*

Specimen	Thermal flux, nvt	Average temp., °F	Thermal cycles	Density, g/cm ³	Dimensional change, in./in.
A-2	2.4×10^{17}	1740	1	2.840	0 ± 0.0007
A-3	3.5×10^{18}	1475	9	2.835	0 ± 0.0007
B-6	4.3×10^{18}	1645	19	2.833	0 ± 0.0007
B-1	6.6×10^{18}	1605	39	2.834	

* Specimens (plates, $\frac{1}{2}$ by $\frac{1}{8}$ by $1\frac{1}{2}$ in.) contained 0.4 wt. % U²³⁵ and generated 120 watts/sq in.

Table III-12 ThO₂-UO₂ SPECIMEN HISTORY*

Specimen	Thermal flux, nvt	Average temp., °F	Thermal cycles	Density, g/cm ³	Dimensional change, in./in.
5	5×10^{18}	1385	13	9.135	0 ± 0.001
6	5×10^{18}	1385	13	9.118	0 ± 0.0008
1	1×10^{20}	1840	33	8.929	0 ± 0.0008
		1475	20		
3	1×10^{20}	1840	33	9.087	Broken in handling
		1475	20		

* Specimens (rods, $\frac{1}{8}$ by $\frac{1}{8}$ by $1\frac{1}{2}$ in.) contained 2.5 wt. % U²³⁵, and the heat dissipation was approximately 125 watts/sq in.

Elements of Low Fuel Content

Uranium-22 Wt. % Zirconium

Initial studies of the effect of heat-treatment on the radiation stability of uranium-22 wt. % zirconium alloy fuel elements have been completed by BMI.¹⁸ The results of these studies are shown in Table III-14.

Fuel Element Studies

Many studies associated with fuel element design, manufacture, or performance are being made. These studies provide useful information to the reactor designer for evaluating the performance of particular fuel elements.

Uranium-Aluminum Alloy Fabrication

Relatively sound homogeneous extrusion billet castings containing up to 30 wt. % uranium have been cast using air melting techniques. Two-in.-diameter right cylinders containing up to 45 wt. % uranium, free of gas porosity and relatively homogeneous, have also been cast.¹⁹

Other studies in conjunction with the casting research has led BMI to conclude the following:

1. The corrosion resistance of aluminum-uranium alloys containing up to 45 wt. % uranium in water at 300°F is comparable to that exhibited by the 16 wt. % uranium alloy. No appreciable difference was noted between the corrosion resistance of as-cast and heat-treated material.

Table III-13 URANIUM LOSS FROM A UO₂-GRAPHITE ELEMENT HEATED IN FLOWING NITROGEN AT 2000°F

Period, hr	Uranium loss rate, g/(cm ²)(hr)
0-28	0.53
28-48	0.57
48-68	0.034
68-93	0.020

Note: Samples consisted of a 30-mil diameter UO₂ rod imbedded in a cylindrical graphite matrix by coextrusion with the graphite mix. The element was heated previously at 2300°F for 4 hr to carbonize the binder.

2. Metallographic studies failed to indicate any diffusion of uranium from the 16 to 45 wt. % uranium alloys into 2S aluminum after 30 days at 750°F.

3. Hot-hardness tests performed on alloys containing from 16 to 45 wt. % uranium indicate that a fabrication temperature above 750°F would be satisfactory for all compositions.

4. Press-forging tests indicated that the alloys in all ranges of composition studies would be amenable to extrusion.

5. The addition of up to 0.5 wt. % silicon to the 16 wt. % uranium alloy made no significant difference in the liquidus-solidus temperatures.

Melting Points of UO₂ and

Other Reactor Refractories

Of interest to designers of nuclear reactors are the thermal properties of some high-temperature ceramics. Table III-15 is a compilation

of some of the results of the Fuels Research Unit at Knolls Atomic Power Laboratory (KAPL) on the determination of high melting points of some ceramics including uranium dioxide.^{20a}

General Properties of Potential Fuel Compounds

There is considerable interest in the refractory binary compounds of uranium as fuel material for moderate- and high-temperature reactors. Table III-16 (reference 21), Table III-17

The conclusions reached by BMI²¹ on their study of powder metallurgical techniques for fabricating some of these fuel elements are listed below.

1. Roll-clad plate type fuel elements may be fabricated by powder-metallurgy techniques to contain up to 30 vol. % of the uranium compounds dispersed in a matrix as indicated in Table III-19.

2. Fabrication methods can be controlled such that discrete particles of the above-named ura-

Table III-14 PRE- AND POSTIRRADIATION DATA ON ZIRCONIUM-22 WT. % URANIUM ALLOY SPECIMENS

Specimen	Pre-irradiation heat-treatment*	Temp. during irradiation, °C	Total calculated burn-up, at. %†	Krypton released, % of theoret.	Density	Length	Diameter	Remarks
EN-19	C	~400	0.95	0.4	-4.06	3.3	1.60	No appreciable bow or warpage is evident
EN-27	E	~400	0.85		-3.28	5.4	-0.30	Small amount of surface roughening; no bow or warpage is evident
EN-4A	F	~400	0.85	0.4	-3.41	7.0	0.00	Machining marks are quite evident; slightly warped
EN-20	C	~400	0.74		-3.41	6.4	2.19	Warped in center section of the specimen
EN-26	E	~400	0.76	4.5	-2.88	10.7	-1.89	Very slight bow is evident
EN-5A	F	~400	0.88	3.5	-3.41	3.2	4.09	Slightly warped
EN-22	C	~400	0.92	0.4	-2.88	4.9	2.89	No bow or warpage is evident
EN-28	E	~400	0.88	0.3	-3.54	5.2	0.10	No bow or warpage is evident
EN-11	F	~400	0.76	0.4	-2.75	2.3	1.29	Slightly bowed
EN-23	C	650	0.16	1.3	-2.75	0.6	0.90	Very smooth surface; no bow or warpage is evident
EN-29	E	650	0.16	0.3	-2.23	-6.8	0.40	Very smooth surface; no bow or warpage is evident; one end of specimen was broken in capsule
EN-12A	F	650	0.13	0.4	-1.97	0.5	0.40	Very smooth surface; no bow or warpage is evident

* The letter denotes the following type of heat-treatment:

C, 36 hr at 1550°F, furnace cooled 1/2°F/min from 1550 to 1200°F and furnace cooled to room temperature.

E, 36 hr at 1550°F, water quenched; annealed 1/2 hr at 1020°F, water quenched; annealed 10 hr at 1400°F and furnace cooled.

F, 36 hr at 1550°F, water quenched; annealed for 22 days at 1020°F and furnace cooled.

† These calculations are based on dosimeter readings.

(reference 20b), and Table III-18 (reference 18) include in them some of the physical and nuclear properties of these materials, along with other pertinent information available.

Fuel Fabrication of Refractory Type Uranium Elements Other than UO₂ and ThO₂

In the program of development of fuel elements for high-temperature reactors, which are also resistant to corrosion and radiation damage, refractory type uranium compounds and fuel elements (other than UO₂ and ThO₂) fabricated by powder-metallurgy techniques are being widely considered. Tables III-16 to III-18 summarize some of the available data on these materials.

niun compounds are dispersed homogeneously in the matrix and such that a good core-to-cladding bond is obtained.

3. UC dispersed in type 302B stainless steel has the best corrosion resistance to NaK of all specimens tested. None of the other combinations was severely attacked.

4. At a 30 vol. % loading, UN in stainless steel is the only core material investigated that appears to have favorable resistance to 680°F degassed water.

5. When a high loading (30 vol. %) of a uranium compound, such as U₂Ti, with poor corrosion resistance in 680°F water is dispersed in Zircaloy-2, little or no change in corrosion resistance occurs with a change in fabrication procedures.

6. It is important to note that only high loadings of dispersed particles were investigated in this program. Other related work with dispersed fuel materials has clearly demonstrated that a substantial improvement in corrosion resistance would be obtained as the volume loading of the highly corrosive fuel dispersant was reduced.

Table III-15 THERMAL DATA^{20a}
L. G. Wisnyi and S. W. Pijanowski

Compound	Atmosphere	Temp., °C	Remarks
UO ₂	H ₂ , He, or A	2750 ± 40	Melting point
Sm ₂ O ₃	H ₂	2300 ± 50	Melting point*
Eu ₂ O ₃	H ₂	2050 ± 30	Melting point*
Gd ₂ O ₃	H ₂	2330 ± 20	Melting point*
Dy ₂ O ₃	H ₂	2340 ± 10	Melting point*
Na ₂ UO ₄	Air	1635 ± 10	Melting point
K ₂ UO ₄	Air	1620 ± 10	Melting point
MgUO ₄	Air	1750 ± 25	Decomposed†
CaUO ₄	Air	1800 ± 10	Stable
SrUO ₄	Air	1800 ± 25	Decomposed†
BaUO ₄	Air	1700 ± 10	Stable
MgUO ₄	O ₂	1700 ± 25	Decomposed†
CaUO ₄	O ₂	1800 ± 10	Stable
SrUO ₄	O ₂	1655 ± 10	Stable
MgUO ₄	He	1540 ± 25	Decomposed†
CaUO ₄	He	1615 ± 35	Decomposed†
SrUO ₄	He	1335 ± 25	Decomposed†
UO ₂ · P ₂ O ₅	Air	1260 ± 10	Melting point
Metallic boron	H ₂	2040 ± 30	Melting point‡

* The melts were composed of some vitreous material and a crystalline state which may be a high-temperature phase or a reduced oxide.

† In all cases O₂ was given off and UO₂ remained in the residue. The alkaline earth is believed to have volatilized since it could not be detected in quantity in the residue.

‡ Residue contained both crystalline and vitreous boron.

7. Good-quality Zircaloy-2 powder may be prepared by the hydride method.

Thoria-base Metallo-ceramic Fuel Elements

Experimental studies have been conducted by members of the Armour Research Foundation and ANL on methods of producing dense thoria (ThO₂) bodies having good thermal shock resistance and adequate mechanical strength and using additives having low thermal-neutron cross sections. The following are the general conclusions as presented in the report.^{20c}

1. Hot-pressing was found to be the only means of producing dense sound fibrous cermet bodies. The resistance to thermal shock and mechanical strength of thoria bodies is vastly improved by the incorporation of small amounts of molybdenum fibers. Thoria bodies with concentrations of molybdenum as low as 5 vol. % show thermal shock resistance adequate for most purposes. Thoria-molybdenum powder cermets show poor resistance to thermal shock.

2. Thoria bodies containing thick fibers of niobium metal possess good resistance to thermal shock and good mechanical strength. Thin fibers of niobium are very prone to carburization which curtails their effectiveness toward imparting good thermal shock resistance to the resulting body.

3. Owing to the fact that zirconium undergoes a phase transition at about 850°C, its fibers and powder become embrittled in hot-pressing, making them unsuitable for incorporation in thoria.

Table III-16 SOME PROPERTIES CONSIDERED IN THE SELECTION OF FUEL COMPOUNDS²¹

Compound	Uranium, wt. %	Density, g/cm ³	Uranium, g/cm ³	Uranium increase, %/given volume	Crystal structure	Melting point, °C	σ_a of second element, barn/atom
UCu ₅	42.8	10.60	4.54	-52.7	FCC	1052	3.69
UNi ₄	44.77	11.31	5.07	-47.1	FCC	1300	4.6
UAl ₃	81.52	8.14	6.64	-30.9	FCC	~1590	0.230
USi ₂ (alpha)	80.91	8.98	7.27	-24.2	BCT	~1600	0.13
USi ₂ (beta)	80.91	9.25	7.48	-22.1	Hex	~1600	0.13
UFe ₂	68.0	13.21	8.98	-6.5	FCC	1235	2.53
UO ₂	88.1	10.96	9.6	0	Cubic	2875	<0.0002
UC ₂	90.83	11.68	10.61	+10.4	BCT	2450-2500	0.0032
UC	95.19	13.63	12.97	+35.4	FCC	2350-2400	0.0032
UN	94.44	14.32	13.52	+40.5	FCC	2650 ± 100	1.88
U ₂ Ti	90.8	15.22	13.8	+43.7	Hex	~890 (decomposition)	5.6
U ₃ Si	96.21	15.58	14.99	+56.2	BCT	930 (decomposition)	0.13
U ₃ Ni	96.05	17.6	16.90	+76.0	BCT	790 (decomposition)	4.6
U ₃ CO	96.1	17.7	17.00	+77.2	BCT	~830 (decomposition)	37.0
U ₃ Fe	96.2	17.7	17.02	+77.5	BCT	815 (decomposition)	2.53
U ₃ Mn	96.2	17.8	17.12	+78.7	BCT	~726 (decomposition)	13.2

Table III-17 AVAILABLE DATA ON REFRACTORY URANIUM COMPOUNDS^{20b}

Compound	Uranium content, g/cm ³	Melting point, °C	Suitable samples prepd.	Reactivity with common elements or compounds	Thermal cond., cal/(sec)(cm ²)(°C/cm)	Thermal expansion coefficient, 10 ⁻⁶ /°C
UC	12.97	2350-2400	In prog.	Lit: Decomposes in water at 83°C*	Lit: 0.078 at 44°C	
U ₂ C ₃	11.96	1775 de-comp.				
UC ₂	10.61	2450-2500	In prog.	Lit: Reacts with water at 82°C; reacts with acids and alkalis Battelle thermodynamic: Expect reaction with oxygen, H ₂ O, steam; no reaction with hydrogen†		Lit: 12.5 at 20-235°C
UN	13.52	2650 ± 100	Partially	Battelle thermodynamic: Expect reaction with oxygen, H ₂ O, steam; no reaction with hydrogen Exptl: Porous compact reacted with acids, boiling H ₂ O; no reaction with 1N NaOH		
US	9.58	>2000	In prog.	Lit: Stable in boiling H ₂ O Battelle thermodynamic: Expect reaction with oxygen, steam		
U ₂ S ₃	7.32					
U ₃ Si†	14.99	930	Yes	Lit: Resists air oxidation up to 200°C. Roughened in lead-bismuth eutectic at 425°C Exptl: Reacts with acids; no reaction with 1N NaOH. Slight weight gain in H at 500°C. Stable in boiling water, slight weight loss in 650°F water. Disintegrated in air at 400°C	Lit: 0.036 at 25°C; 0.041 at 50°C	Exptl: 13.0 at 20-200°C; 17.5 at 20-750°C
U ₃ Si ₂	11.31	~1650	Yes	Exptl: In progress		Exptl: 15.5 at 20-200°C; 14.6 at 20-950°C
USi	9.30	~1600	Yes	Exptl: In progress		
USi ₂	7.27	~1600	Yes	Exptl: Reacts with acids; no reaction with 1N NaOH. Slight weight gain in H at 500°C. Stable in boiling water. Slight weight gain in air at 400°C	Exptl: In prog.	
α USi ₂	7.48	~1600				
β USi ₃	6.02	~1500	Yes	Exptl: In progress	Exptl: In prog.	Exptl: 13.4 at 20-200°C; 16.3 at 20-950°C
UB ₂	11.75	>1500	In prog.			
UB ₄	7.94	>1500	In prog.			
U ₆ Ni	16.90	790	Yes	Exptl: In progress		
UNi ₂	9.02	985	Yes	Exptl: In progress		
UNi ₅		1300				
UAl ₂	6.64	~1590	Yes		Exptl: In prog.	
U ₆ Fe	17.00	815				
UFe ₂	8.98	1235				

* Lit: Information from the literature.

† Exptl: Experimental data from Battelle investigation. Battelle thermodynamic: estimates made at Battelle.

‡ Ultimate strength, psi: Lit., 37,000 tension; 280,000 compression.

4. The thermal conductivity of thoria is somewhat increased by the incorporation of molybdenum fibers.

5. The presence of metal fibers within thoria has the effect of lowering Young's modulus of

nesium into zirconium embrittled the zirconium but did not change the appearance of the microstructure. The data presented would be applicable to uranium-magnesium dispersion elements which are canned in zirconium.

Table III-18 PROPERTIES OF SOME SINTERED URANIUM COMPOUNDS¹⁸

Compound	Sintering temperature		Bulk density		Linear thermal expansion coefficient, $10^{-6}/^{\circ}\text{C}$			
	$^{\circ}\text{F}$	$^{\circ}\text{C}$	g/cm^3	% of theor.	Heating		Cooling	
					20 to 100°C	20 to 950°C	20 to 100°C	20 to 950°C
U_3Si_2	2820	1550	12.0	98.0				
USi	2950	1620	9.8	94.0	20.0	15.7	18.8	17.3
USi_2	2840	1560	9.0	97.5	15.9	16.9	15.1	16.7
USi_3	2730	1500	7.6	93.0	15.5	14.9	15.5	19.1
UAl_2	2460	1350	7.15	88.0	14.9	16.2	15.2	17.1

elasticity for the body. Molybdenum fibers produce this effect in a more pronounced manner than niobium fibers owing to the embrittlement of the latter in hot-pressing. Niobium powder does not perceptibly affect the modulus of elasticity value of thoria. Young's modulus of elasticity value of various thoria bodies dropped sharply with one thermal shock. After the third thermal shock this value for thoria-molybdenum fibrous cermets remained fairly constant. Although the modulus of elasticity value of the thoria-niobium fibrous cermets remain high after the first few thermal shocks, the samples show signs of breaking after the fifth such shock.

6. The theoretical density value of $10.0 \text{ g}/\text{cm}^3$ for thoria based on present X-ray study is higher than the value frequently encountered in the literature.

7. X-ray diffraction studies revealed that niobium metal in contact with graphite during hot-pressing at 1500°C becomes carburized. The carbide formed is not the known phase NbC , but an unknown one with less carbon. Molybdenum liners considerably reduce the amount of carburization.

Magnesium-Zirconium Diffusion

An investigation has been made by KAPL on the rates of diffusion, or solid-state reaction, in magnesium-zirconium couples.²² The interdiffusion rates between magnesium and zirconium were determined for the temperature range of 500 to 640°C . The diffusion of mag-

Uranium-Aluminum-Silicon

Eutectic Alloy Diffusion

Extensive use has been made, in the manufacture of uranium fuel elements, of an aluminum-silicon eutectic (AlSi) as a bonding material between the fuel and the aluminum or aluminum alloy jacket. The use of such fuel elements, especially in water cooled reactors, is limited by the corrosion resistance of the jacket material as well as the possible penetration of the jacket material by the solid-state diffusion of uranium through the bond and then the aluminum alloy jacket. Hanford has recently completed a study of the diffusion of uranium and AlSi eutectic alloy.²³ The maximum rates of U/AlSi diffusion were determined at temperatures of 200 , 250 , and 300°C . The maximum penetration was found to have the following approximate relation:

$$x = (K_0 t e^{-Q/RT})^{1/2} = (K_2 t)^{1/2}$$

where x = maximum over-all penetration

t = time

T = absolute temperature

K_0 = constant

Q = activation energy (assumed constant over temperature range)

R = gas constant

$K_0 = 9.7 \times 10^{10} \text{ mils}^2/\text{hr}$ for diffusion of uranium into AlSi

$Q = 3.15 \times 10^4 \text{ cal/mole}$ for diffusion of uranium into AlSi

$K_0 = 6.75 \times 10^9$ mils²/hr for diffusion of AlSi into uranium

$Q = 2.97 \times 10^4$ cal/mole for diffusion of AlSi into uranium

Although one expects irradiation to affect the rate of solid-state diffusion, it was pointed out

extraction, using tributyl phosphate and hexone, was used for recovery of the uranium.

Fuel Element Rupture Tests

A summary report of fuel element rupture tests in high-temperature, high-pressure recirculating water systems has been prepared

Table III-19 COLD-PRESSED DENSITIES OF COMPACTS PRESSED AT 50 TSI

Fuel	Matrix	Theoretical density of mixture, g/cm ³	Cold-pressed* density, g/cm ³	Theoretical density, %
U ₂ Ti	Zircaloy-2	9.15	7.32	80.0
U ₂ Si	Zircaloy-2	9.25	7.30	79.0
U ₆ Ni	Zircaloy-2	9.85	7.60	77.2
UC	Zircaloy-2	8.67	6.80	78.5
UC	Type 302B stainless steel	9.62	7.56	78.6
UN	Type 302B stainless steel	9.82	7.63	77.7
UN	Type 18-8 elemental powder stainless steel	9.82	7.77	79.2

* Average values.

that the rate of solid-state diffusion between various metals under irradiation did not change materially. The data obtained in this study will be used as a basis for interpreting the results obtained on the diffusion rates of irradiated specimens.

Metallic Impregnation of UO₂

Several studies have been previously reported on additives to UO₂ and ThO₂ fuels to improve their corrosion, thermal-fracture, and thermal conductivity.^{1,24,25} These studies consisted of additions, by powder metallurgy techniques, of beryllia, beryllium, ceria, silica, silicon, and zirconia, as well as metal fibers of molybdenum, niobium, and zirconium.

Processing the First Zirconium Fuel Core

Processing of the uranium-zirconium fuel from the first Submarine Thermal Reactor (S1W) core has been accomplished at the Idaho Chemical Processing Plant.²⁶ Because the uranium-zirconium alloy fuel does not dissolve in the nitric acid system used at the Idaho Plant, a head-end process using hydrofluoric acid was substituted to dissolve the elements. Solvent

by Hanford.²⁷ Uranium is known to corrode fairly rapidly in water, the rate of corrosion increasing with water temperature. Thus a failure of a fuel element can result in the uranium corroding and swelling to the point where a rupture of the process tube in which the element is located occurs. This in turn can cause severe damage to the rest of the reactor, especially in graphite moderated reactors.

The study reported in the reference report describes a series of simulated rupture tests performed in a high-temperature, high-pressure recirculating loop to determine the effects of simulated pinhole ruptures of fuel elements of various designs. The gross effects related to process tube plugging were studied, and quantitative information on tube plugging rates related to various coolant temperatures were obtained.

Erosion of UO₂ Pellets

Testing of bare, cold-pressed and sintered UO₂ pellets in 525°F, 2000 psia water having an oxygen content less than 0.1 cm³/kg, a pH between 9.5 and 10.5 and flow velocities of 20 ft/sec showed no significant erosive attack on UO₂ pellet surfaces. Exposures were for 183 hr, one specimen being exposed for 397 hr.²⁸

Corrosion of Defected Zirconium-clad

UO₂ Fuel Rods

Tests studying the rate of corrosive attack on defected PWR fuel rods in oxygenated water have been completed at Bettis.²⁰ Corrosion was limited to localized areas beneath the defects and was in the form of small pitting and some discoloration. Weight losses of pellets under defects were not great. In a 154-hr run bare pellets showed a weight loss 84 times the weight loss of the most severely attacked pellet in a fuel rod; in another run of 137 hr, 12 times as great a weight loss.

Water conditions were 600°F, pH 9 to 10 with LiOH, 3 to 25 cm³ O₂/kg H₂O and a water velocity of 17 ft/sec. The results indicate that UO₂ pellets in defected fuel rods do not experience a rapid corrosive attack when exposed to oxygenated high pH water despite the high rate of attack on UO₂ exposed within cladding in this water condition.

Briefly Noted Reports

A comprehensive summary coverage of the mechanical, physical, chemical, and metallurgical properties of uranium and its alloys has been made by the British in a recent magazine article.³⁰ Selected values for the above properties are given and the effects of irradiation, thermal cycling, fabrication history, and alloy additions are discussed. Some of the alloys discussed are U-Mo, U-Cr, U-Zr, U-Ti, U-V, U-Th, U-Ge, U-Fe, and U₃Si. A useful data sheet is presented containing summary tables of the physical properties; mechanical properties; compatibility with other metals, gases, and liquids; and irradiation effects of uranium.

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Control Materials

Among the rare earths, europium, gadolinium, samarium, and dysprosium are potential control materials for use in nuclear reactors. Europium, in particular, because of the high cross-section neutron-born daughters, is being studied^{1,2} as a control material for long-lived reactor cores. Table IV-1 lists some of the control materials.

Of these materials, cadmium absorbs strongly only in the thermal energy range and hence is not so effective as the other materials in reactors such as H₂O moderated reactors which have relatively high epithermal flux components. Boron, a 1/v absorber, is quite effective in the epithermal range if it is used in sufficiently thick and sufficiently concentrated sections. The rare earths have strong resonances in the epithermal energy range and are therefore also effective as epithermal absorbers.

Satisfactory techniques have been developed for fabricating stainless-steel clad Eu₂O₃* Y-shaped control rods.³ The core material was prepared by powder-metallurgy techniques as dispersions of 25, 30, and 40 vol. % of Sm₂O₃ in type 302B stainless-steel powder and hot coextruded with a type 304 stainless-steel can.

Boron Compounds

A control rod of high effectiveness can be made by using boron as the control material.

Impact and tensile specimens of 1 wt. % B¹⁰, 2 wt. % B¹⁰, and 5 wt. % B_{nat} stainless-steel powder dispersions were irradiated in MTR to a maximum unperturbed exposure⁴ of 1×10^{21} nvt. The results showed no gross dimensional changes or warpage as a result of irradiations. However, almost complete embrittlement occurred with exposures as low as 0.8×10^{20} nvt, resulting in a large (factor of 10 to 20) decrease in the impact strength of the material.

Clad platelet specimens containing cores of titanium-5.0 wt. % B¹⁰ dispersion material have

been irradiated to a thermal-flux exposure⁵ of 1.97×10^{21} nvt. These specimens underwent gross core volume expansion which resulted in rupture of the clad. The same material exposed to 9.5×10^{20} thermal nvt had only a volume increase of 3 per cent, and specimens exposed to a thermal flux of approximately 4×10^{20} nvt suffered no significant volume increase.

Table IV-1 CONTROL MATERIALS

Element	Density, g/cm ³	σ_a , barns	Material	Cost, \$/lb
B _{nat}	2.331	750	Ferroboron (17.5% B _{nat})	1.20*
Cd	8.6	2,400	Cadmium	1.70*
Hf	13.3	115	Hafnium	125.00*
Sm	6.9	5,500	Sm ₂ O ₃	113.00†
Eu	5.2	4,600	Eu ₂ O ₃	2,270.00†
Gd	7.9	46,000	Gd ₂ O ₃	408.00†
Dy	8.5	1,100	Dy ₂ O ₃	2,720.00†

*Price from Steel, May 1954, market prices.

†Price from Nucleonics, January 1957, p. 44.

Irradiation tests have been conducted on Zircaloy-2 clad zirconium-boron specimens for use as discrete burnable poison elements.⁶ The following compositions were tested:

1. B¹⁰ (0.21 wt. %) alloyed with Zircaloy-2
2. B¹⁰ (0.35 wt. %) alloyed with Zircaloy-2
3. B_{nat} (1.2 wt. %) alloyed with Zircaloy-2
4. B¹⁰ (0.35 wt. %) dispersed in zirconium

The poison materials were metallurgically bonded to the Zircaloy-2 cans and irradiated in the MTR in a thermal flux of 2×10^{14} nv to over 90 per cent burn-up of poison atoms. All the test elements exhibited varying degrees of surface damage, blisters, spalled areas, and clad cracks. It was concluded that metallurgically bonded elements of this type were unsatisfactory but that further studies should be conducted with discrete burnable poisons with unbonded claddings.

Hafnium-Boron

The use of hafnium as a control-rod material because of its excellent adsorption and burn-out characteristics is limited only by its cost and

*Sm₂O₃ was actually used in the fabrication studies for economic reasons and because of its physical and chemical similarity to Eu₂O₃.

availability. Since in most reactors only a small section of the control rod lies in a high-flux region of the reactor, it would be desirable that a control rod be made of a composite of hafnium in the high-flux end of the rod and a relatively inexpensive material, but of high thermal neutron absorption cross section, in the rest of the rod.

Moderator, Shielding, and Structural Materials

Moderator Materials (Graphite)

A summary of the irradiation data of graphite⁷ has already been published in *Power Reactor Technology*, Vol. I, No. 1, December 1957. In addition, fundamental studies on several commercial and experimental graphites to determine the relation between internal structure and dimensional stability under irradiation have been carried out at Battelle.⁸ The results of this investigation indicate that the net dimensional change obtained at a given irradiation temperature is dependent on two competing effects: (1) crystal expansion and (2) shrinkage of intercrystalline voids.

It was further determined that at room temperature the rate of crystal expansion is high and commercial graphites expand considerably under irradiation. However, graphites whose structures contain the proper amounts of intercrystalline micropores are dimensionally stable under the same conditions.

At temperatures of 350 to 450°C, the rate of crystal expansion is reduced under irradiation, and shrinkage of intercrystalline voids causes a net contraction of bulk graphite. The report⁸ concludes that graphite with a minimum of intercrystalline porosity should be used at the elevated temperatures.

Zirconium and Zirconium Alloys

The mechanical properties of zirconium and zirconium alloys at elevated temperatures are important because of their extensive application to the power reactor field. Of particular importance are the burst strengths of tubing, the tensile strength, creep strength, and corrosion properties of these materials.

1. *Physical Properties.* A fairly detailed study on the properties of zirconium and zir-

conium alloys is being conducted by BMI, and some of the results to date follow.⁹⁻¹²

Table IV-2 is a compilation of representative tests on the burst strengths of zirconium and Zircaloy-2 tubes at temperatures of 75, 300, 450, and 650°F. Tests on tubes which failed because of a defect are not included in the table.

Table IV-3 contains the latest data on the creep properties of annealed Zircaloy-2 and Zircaloy-3A at elevated temperatures and under various stresses. Deformation under load has been checked for times of 1000 hr and in a number of cases up to 3000 hr.

Table IV-4 includes a compilation of the mechanical properties of annealed and 35 per cent cold-worked Zircaloy-3A at elevated temperatures.

2. *Corrosion Studies.* Long-term corrosion studies on Zircaloy-2 in 600 and 680°F degassed water indicate excellent corrosion resistance after approximately 3½ years of exposure. The following data are reported on intentionally contaminated specimens of Zircaloy-2, exposed to 680°F degassed water for 23 months.^{10,11}

1. Nitrogen levels in excess of 200 ppm result in an almost complete loss of corrosion resistance.

2. Hydrogen in the range 500 to 1500 ppm does not significantly affect the corrosion behavior of Zircaloy-2.

3. A substantial increase in corrosion rates occurs with oxygen additions in excess of 6000 ppm.

As part of a program to develop zirconium-base alloys superior to Zircaloy-2, alloys of zirconium containing 0.25 to 0.50 wt. % tin plus 0.03 to 0.50 wt. % iron and/or nickel with and without additions of nitrogen and aluminum have been corrosion-tested in 680°F degassed water and 750°F steam. After exposures up to 616 days, the results as reported to date indicate the following.¹⁰⁻¹²

1. Heat treatments consisting of either a slow cool from 775°C or a water quench from 900°C do not significantly alter the corrosion behavior of tin-iron or tin-nickel alloys.

2. Sponge-base alloys containing 0.2 to 0.5 wt. % tin plus 0.03 to 0.5 wt. % nickel are more resistant to 750°F steam than are alloys containing similar combinations of tin and iron. The reverse, however, is true for 680°F water exposure.

Table IV-2—BURST STRENGTHS OF ZIRCONIUM AND ZIRCALOY-2 TUBES AT ELEVATED TEMPERATURES*

Material	Outside diameter, in.		Wall thickness, in.		Cold work, %	Rockwell hardness	Burst pressure, psi	Burst stress, [†] psi	Circumferential elongation, [‡] %	Remarks
	Nominal	Measured	Nominal	Measured						
Room temperature										
Zirconium	1.5	1.500	0.060	0.060	0	75.0-B	6,750	77,680	21.9	
Zirconium	1.5	1.502	0.060	0.061	40	90.0-B	9,400	105,990	15.8	
Zirconium	1.5	1.510	0.020	0.0250	10		3,900	113,650	23.1	
Zirconium	1.5	1.505	0.020	0.0250	10		3,850	112,190	17.1	
Zirconium	1.5	1.507	0.060	0.065	10		10,000	105,795	38.6	Fractured in weld
Zirconium	1.5	1.506	0.060	0.0648	10		9,900	104,635	31.8	Fractured in weld
Zircaloy-2	1.5	1.505	0.035	0.0385	10		5,300	99,520	17.8	
Zircaloy-2	1.5	1.503	0.035	0.0380	10		5,600	105,221	16.6	
300°F										
Zirconium	1.5	1.505	0.060	0.060	0	75.0-B	4,650	53,380	42.1	
Zirconium	1.5	1.502	0.060	0.060	40	90.0-B	6,800	78,085	16.4	
Zirconium	1.5	1.505	0.020	0.0250	10		3,200	93,060	30.5	
Zirconium	1.5	1.506	0.020	0.0250	10		3,200	93,250	12.9	
Zirconium	1.5	1.506	0.060	0.065	10		6,650	70,335	2.1	Small hole in weld
Zircaloy-2	1.5	1.504	0.035	0.039	10		4,750	87,715	18.6	Fractured in weld
Zircaloy-2	1.5	1.503	0.035	0.038	10		4,750	89,125	18	Fractured in weld
450°F										
Zircaloy-2	0.5	0.499	0.035	0.0348	10-15	35.0-30N	8,500	50,410	54.5	
Zircaloy-2	0.625	0.625	0.049	0.0490	10-15	35.0-30N	9,700	51,540	29.0	
Zirconium	1.5	1.506	0.060	0.065	10		5,700	60,224	3.3	Small hole in weld
650°F										
Zirconium	1.5	1.499	0.020	0.020	0	16.1-30N	1,100	40,150	32.4	
Zirconium	1.5	1.495	0.020	0.020	40	32.2-30N	1,200	43,740	17.4	
Zirconium	1.5	1.501	0.060	0.060	0	75.0-B	2,500	28,750	44.2	
Zirconium	1.5	1.501	0.060	0.060	40	90.0-B	4,600	52,940	17.4	
Zircaloy-2	1.5	1.495	0.020	0.020	0	15.4-30N	1,650	60,260	37.0	
Zircaloy-2	1.5	1.500	0.020	0.019	40	35.2-30N	1,650	63,655	17.0	
Zircaloy-2	1.5	1.496	0.035	0.036	10-15	86.9-B	2,600	51,385	34.5	
Zircaloy-2	1.5	1.502	0.060	0.060	0	79.7-B	3,600	40,090	57.0	
Zircaloy-2	1.5	1.502	0.060	0.061	40	93.8-B	6,050	63,025	20.0	
Zircaloy-2	0.5	0.499	0.035	0.0343	10-15	35.0-30N	7,500	45,965	45.5	
Zircaloy-2	0.625	0.625	0.049	0.049	10-15	35.0-30N	7,900	41,975	29.1	
Zirconium	1.5	1.502	0.020	0.025	10		2,000	58,320	41.8	Fractured, not in weld
Zirconium	1.5	1.506	0.060	0.065	10		4,700	50,213	37.3	Fractured, not in weld
Zircaloy-2	1.5	1.502	0.035	0.038	10		3,050	57,229	26.8	Fractured in weld

*All data obtained from Reports BMI-1173, 1181, 1201, 1213, and 1220.

[†]Calculated by Barrow's thin-wall tube formula, $S = Pd/2t$, where S is hoop stress, P is internal pressure, d is inside diameter, and t is wall thickness.[‡]Based on change in diameter at fracture point.

Table IV-3 CREEP PROPERTIES OF ANNEALED ZIRCALOY-2 AND ZIRCALOY-3A*

Deformation at indicated time, %											
Temp., °F	Stress, psi	Load on	100 hr	500 hr	1000 hr	1500 hr	2000 hr	3000 hr	Total	Load off	Creep rate, %/hr*
Zircaloy-2											
300	23,000	0.39	0.97	1.09	1.15						0.00012
300	23,000	0.20	2.20	2.52	2.63						0.00023
300	23,000	0.175	1.96	2.29	2.39						0.00020
450	21,000	0.48	0.93	0.96	0.98						0.00040
450	21,000	0.59	2.465	2.49	2.49	2.495	2.495		2.495	2.37	< 0.00001
450	23,000	0.30	2.05	2.13	2.15						0.00003
450	23,000	1.85	3.90	3.93	3.95						0.00005
450	24,500	0.90									
450	24,500	2.40	9.90	10.25	10.25	10.25					< 0.00001
550	17,500	0.50	1.15	1.337	1.65						0.00063
550	22,500	2.65	4.62	4.64	4.65	4.658	4.658		4.658	3.679	< 0.00001
550	24,500	7.50	16.00	16.10	16.10						< 0.00001
650	9,000	0.032	0.072	0.072	0.81	0.090	0.091	0.091	0.091		< 0.00001
650	9,000	0.06	0.1	0.117	0.117	0.117	0.117	0.117	0.117	0.085	< 0.00001
650	12,500	0.17	0.428	0.448	0.46				0.46		0.000024
650	12,500	0.15	0.245	0.282	0.335	0.342	0.350	0.380	0.385		0.00003†
650	17,500	0.54	0.675	0.711	0.742						0.00006
650	17,500	1.385	1.55	1.594	1.63	1.65	1.686		1.705		0.000056†
650	17,500	1.37	1.615	1.707					1.707		0.000047
650	21,000	1.50	3.89	4.06	4.36						0.003
650	22,500	6.0	7.00	7.35	7.92				8.02	7.90	0.0008
Zircaloy-3A											
300	25,000	0.25	3.37	5.05	6.05				6.35	6.07	0.0017
475	18,500	1.0	2.16	2.17	2.19				2.19	1.94	0.00002
475	22,500	2.3	17.0	23.4	25.75						0.003
475	20,000	0.5	2.68	2.70	2.72						0.0004

*Based on creep deformation between 500 and 1000 hr.

†Based on creep deformation between 1000 and 2000 hr.

Table IV-4 MECHANICAL PROPERTIES OF ANNEALED AND 35 PER CENT COLD-REDUCED ZIRCALOY-3A

Material	Temp., °F	Elastic modulus, 10 ⁶ psi	0.2% offset yield strength, 10 ³ psi	Ultimate tensile strength, 10 ³ psi	Elongation, %	Reduction, in area, %
35% cold-worked transverse	79	15.3	92.0	97.7	16.0	45.4
	212	14.5	80.2	83.5	15.5	50.5
	302	12.3	68.0	70.7	15.0	54.0
	392	12.1	61.7	64.8	16.0	60.4
	482	9.8	54.5	55.9	13.0	
Longitudinal Annealed	392	9.85	61.9	65.0	15.5	46.5
	79	14.4	56.7	68.5	32.8	48.6
	79	14.6	57.8	68.5	36.0	52.7
	212	13.6	41.7	50.7	43.0	57.7
	302	14.0	32.4	41.7	54.0	55.9
transverse	392	13.4	24.3	33.1	56.0	60.5
	482	11.3	18.3	29.3	53.0	70.8
Longitudinal	79	14.3	54.2	69.6	30.0	48.0
	392	12.6	22.6	35.1	53.0	43.1

3. As the nitrogen content is increased from 70 to 300 ppm, there is a progressive increase in the corrosion rate of 0.25 to 0.5 wt. % tin-0.25 to 0.50 wt. % iron alloys.

4. The modified Zircaloy-2 alloys show a decrease in corrosion resistance with aluminum contents of 200 ppm.

Pronounced improvement has been noted⁵ in the corrosion resistance of Zircaloy-3 which has been subjected to a laboratory thermal treatment of 1880°F for ~8 hr. The discontinuous white stringers and dull surface condition were absent, but an adherent, lustrous, black oxide was observed.

3. *Welding.* A study has been completed by KAPL on the effect of welding atmosphere and pickling on the corrosion resistance of welded Zircaloy-2 and Zircaloy-3 subjected to 680°F water.¹³ As a result of experiments it was concluded that the best corrosion-resistance welds are obtained with the highest purity gas shielding; the effect of atmosphere is more pronounced in welds made with high heat inputs; repickling after welding does not improve corrosion resistance of welds made in the purest atmosphere but does improve corrosion resistance of welds made in poor atmosphere; the repickling solution, however, tends to become trapped in the crevices formed by backing pads and causes increased corrosion.

4. *Brazing.* Some success has been achieved in the nickel-eutectic brazing of Zircaloy-2 plate type fuel elements at Bettis.¹⁴ Strength and corrosion properties of the joints were reported to be good.

For pure nickel deposits it was found that the optimum brazing conditions involve 0.0003- to 0.0004-in.-thick deposits, rapid heating of assembled components to a brazing temperature of 1000°C to minimize diffusion of nickel prior to eutectic melting, and an extended brazing time of 1 hr in vacuum to diffuse Zircaloy-2-nickel from the joint interface. Improved billets resulted from the substitution of a chemically deposited 0.00022-in.-thick layer of nickel-10 wt. % iron-2 wt. % chromium for a similar thickness of pure nickel, apparently because the alloy deposit is less subject to diffusion on slow heating.

Corrosion testing of brazements prepared so that the Zircaloy-2-nickel is diffused away from the joint was made in 680°F water for 127 days.

Specimens exhibited the normal behavior of Zircaloy-2. Those combining a continuous Zircaloy-2-nickel phase show oxide penetration at the joint of 5×10^{-6} in./hr.

This method shows excellent promise as a means of fabricating low-cost compartmented plate type fuel elements, according to the referenced information.

Sample brazements using 5 per cent beryllium-Zircaloy brazing alloy have successfully passed pressure, thermal cycling, and deformation tests.⁵ It is stated that the brazing technique is well established, but a token effort is still being expended to find a more ductile brazing alloy. No information was given on the corrosion resistance of the brazing alloy.

Zirconium Hydrides

Many studies have been made on the production of hydrogenous material for use as a moderator or radiation shield for nuclear reactors. In this respect good moderation and gamma attenuation are obtained by the combination of hydrogen with a heavy metal, provided the neutron absorption of the heavy metal is low. Hydrides used as radiation shields must use containers that are impervious to hydrogen and capable of withstanding high pressure. General data on various hydrides, including zirconium hydrides, can be found in reference 15.

BMI reports,¹⁶ in their study of core materials for gas-cooled reactors, the following tensile strengths of $ZrH_{1.80}$.

Temperature, °F	Tensile strength, psi	Elongation, %
1130	9700	0
1275	4060	0

Vanadium and Vanadium Alloys

Among the various metals being studied as possible structural and canning materials, vanadium and vanadium alloys show some promise as fuel-element jacket materials for high-temperature sodium-cooled reactors.¹⁷

Table IV-5 compares a few of the properties of uranium and some of the more common high-temperature canning materials.

Where high fuel content is required, the use of metallic instead of ceramic fuel is usually

preferred. However, a limitation to the use of metallic fuels is the low melting point of the eutectic composition of uranium with clad materials such as stainless steel. At the interface of a fuel element consisting of uranium mechanically bonded to stainless steel, the iron-uranium eutectic will melt at temperatures above 725°C. This puts a maximum limit of 725°C on the

sents a basic problem on chemical processing of reactor fuels. It has been found that the corrosion of type 309 Cb stainless steel in mixtures of 2M to 10M HNO₃ and 0.01M to 1.5M HF is much less than that of type 304L stainless steel.¹⁸ The difference in corrosion rate decreases with increasing temperature. The corrosion rate was approximately a linear

Table IV-5 PROPERTIES OF SOME HIGH-TEMPERATURE STRUCTURAL MATERIALS

Material	Density, g/cm ³	Melting point, °C	Thermal neutron cross section		Thermal cond. (k), cal/(sec)(cm)(°C)
			σ_a , barns	$\Sigma_a \times 10^3$, cm ⁻¹	
U	18.4-19	1133	7.4		0.060 @20°C
Zr	6.50	1845	0.180	7.65	0.039 @100°C
Nb	8.57	2415	1.1	61.0	0.12 @100°C
Mo	10.2	2622	2.5	160.0	0.32 @20°C
347 SS	7.98	~1400	~3.80	~246.0	0.037 @100°C
Cr	7.19	1850	2.9	241.0	0.16 @20°C
Ni	8.9	1455	4.6	420.0	0.22 @25°C
V	6.1	1900	4.98	360.0	0.074 @100°C
Ti	4.5	1670	5.6	317.0	0.41 @25°C

temperature of the coolant. Vanadium and uranium form a eutectic melting at 1040°C which would allow some 300°C higher operating temperature.

Information¹⁷ on the tensile, yield, and creep strengths of various vanadium alloys indicates that most of these alloys have higher strengths than type 347 stainless steel. Binary alloys of vanadium containing 5, 10, and 20 wt. % titanium have a 100-hr rupture life at ~50,000 psi and 650°C as compared to ~27,000 psi for type 347 stainless steel. This would indicate that very thin jackets could be used.

The data on corrosion resistance of the vanadium alloys indicate that they are generally corrosion resistant to sodium, at least up to 700°C, when Na₂O is kept down to 10 ppm in the system and are nonresistant to degassed water at elevated temperatures.

The thermal conductivity of vanadium is high compared to 347 stainless steel and is roughly equivalent to it when alloyed with as much as 30 per cent titanium.

Stainless Steel

The fact that the addition of a small amount of hydrofluoric acid to nitric acid to promote dissolution of fuel materials also corrodes the stainless-steel container at a rapid rate repre-

sents a basic problem on chemical processing of reactor fuels. It has been found that the corrosion of type 309 Cb and 304L stainless steel were 44 and 204 mils/year, respectively.

Aluminum

1. *Corrosion.* In a study of the corrosion of aluminum in high-temperature water, KAPL has made a series of tests using sodium silicate as a corrosion inhibitor.¹⁹ The results of the tests indicated that 2S aluminum exposed to 0.1 to 1 per cent solutions of sodium silicate at 200°C showed a marked improvement in corrosion resistance. At 250°C, the corrosion rate increases exponentially with time in the above solution. However, the rate of corrosion is approximately 1/100 of that of pure water. After 350 hr exposure the attack by the silicate solution is about the same as that of pure water after 24 hr exposure.

In addition to previous data ANL has made further tests on the corrosion of aluminum alloys at 290°C. Table IV-6 summarizes the data reported in reference 20.

2. *Fabrication.* An ultrasonic welding technique was developed by AeroProjects, Inc.,²¹ for bonding small aluminum ribs to the outside surface of aluminum or aluminum-clad tubing.

The ribs can be attached at a 12- to 30-in. spiral pitch on tubes 16 to 18 ft long, having a wall thickness of $\frac{1}{8}$ in. or greater and a diameter of 1.9 to 3 in. Tubing with a wall thickness of 0.07 in. required a mandrel backup.

Table IV-6 CORROSION OF ALUMINUM ALLOYS AT 290°C

Alloy*	Solution	Rate, mdd	Intercept, mg/dm ²
M388 (0.9 Ni - 0.3 Fe)	H ₃ PO ₄ (ph 4.5 at R.T.)	1.5	30
M388	Mg ₃ (PO ₄) ₂ (~4 ppm PO ₄ ⁻³)	3.7	37
M388	MgHPO ₄ (~5 ppm PO ₄ ⁻³)	1.3	28
A198x (1 Ni - 0.1 Ti)	Distilled H ₂ O	1.4	148
A203x (5.6 Ni - 0.3 Fe - 0.2 Ti)	Distilled H ₂ O	1.6	160

* Alloy compositions are given in weight per cent.

Miscellaneous

A summary of 6 years experience with wear and friction of materials in water environments of the pressurized-water nuclear power reactor type has been made by Bettis Plant.²² The materials tested are typical of those used in control-rod drive mechanisms, valves, and pumps.

Reactor Coolants: Liquid Metal

A comparison of the properties of liquid metals, Table IV-7, shows that lithium has good potential as a reactor coolant material. Compared to sodium it has a higher boiling point, lower density, and greater heat capacity, making it superior to sodium as a heat-transfer medium. Assuming that the 92.4 per cent abundant isotope Li⁷ can be produced in a pure form, then this coolant would have a macroscopic cross section one-tenth that of sodium. Activation of lithium is also less, since the only induced activity will be due to Li⁸ which is a 0.83-sec beta emitter.

The major deterrent is the high corrosiveness of lithium to containers made of the more common materials such as stainless steel.

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2. W. K. Anderson, Rare Earths Show Promise as Reactor Control Materials, *Nucleonics*, 15(1): (January 1957).
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Table IV-7 PROPERTIES OF SOME LIQUID METALS

Liquid metal	Σ_a ,* cm ⁻¹	Mp, °C	Bp at 760 mm Hg, °C	Density, g/cm ³	Sp. heat, cal/g/°C	Viscosity, CP	Thermal cond., cal/(sec)(cm)(°C)
Li	2.95	179	1317	0.5(200°C)	1.4(200°C)	0.54(216°C)	0.09(225°C)
Li ⁶ (7.52%)	39.0				1.0(500°C)		
Li ⁷ (92.40%)	0.00144						
Na	0.012	97.8	883	0.928(100°C)	0.32(200°C)	0.686(103°C)	0.20(100°C)
					0.30(600°C)	0.182(700°C)	0.16(500°C)
Na-44 wt. % K	0.0185	19	825	0.886(100°C)	0.27(100°C)	0.546(103°C)	0.06(100°C)
					0.25(600°C)	0.161(700°C)	0.07(500°C)
Bi	0.00093	271	1477	10.0(300°C)	0.034(271°C)	1.662(304°C)	0.041(300°C)
					0.039(800°C)	0.996(600°C)	0.037(700°C)
Pb-55.5 wt. % Bi	0.00286	125	1670	10.46(200°C)	0.035(144-358°C)	1.7(332°C)	0.023(200°C)
						1.17(600°C)	0.027(320°C)
Ga	0.15	29.92	1983	6.0(324°C)	0.082(12-200°C)	1.89(52.9°C)	0.07(30°C)
						0.81(500°C)	
Mg	0.0023	651	1103	1.57(651°C)	0.317(651°C)		
					0.337(1027°C)		
Al	0.012	660	2450	2.38(660°C)	0.259(660-1000°C)	2.9(700°C)	0.247(700°C)
						1.4(800°C)	0.290(790°C)
Hg	14.7	-38.87	357	13.64(-20°C)	0.033(0°C)	1.85(-20°C)	0.019(0°C)
					0.032(450°C)	1.01(200°C)	0.03(220°C)

* Macroscopic absorption cross section at melting point.

4. J. D. Eichenberg, The Effects of Irradiation on the Properties of Boron-Stainless Steel Powder Dispersion Samples, WAPD-160, January 2, 1957. (Unclassified AEC report.)
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21. J. B. Jones, C. F. DePrisco, et al., Attachment of Spiral Ribs to Aluminum Tubes by Ultrasonic Welding, DP-168, January 1957. (Confidential AEC report.)
22. N. B. Dewees, B. Levy, and I. A. Marsh, Paper No. 1: Wear and Friction of Materials in Room Temperature and 500°F High Purity Water; taken from Bettis Technical Review, Vol. 1, No. 2, Reactor Metallurgy, WAPD-BT-2, July 1957. (Unclassified AEC report.)

There has been a proposal¹ by the HAPO of the General Electric Company for a program of research and development aimed at the economic recycling of plutonium in power reactors. The program involves an expenditure at the rate of \$4,000,000 per year and the construction and operation of a special developmental reactor. The reactor is estimated to cost \$14,300,000.

The proposal points out that the present price schedule for uranium and plutonium encourages reactor operators to burn uranium and sell plutonium to the government but that as the price of plutonium is allowed to seek its level as a reactor fuel the economic incentive for recycling will develop. It is proposed that meanwhile development of recycle technology should be undertaken because of the long-term advantages of such operation. These advantages are stated to be the following:

1. Unless plutonium is recycled, large stocks of the material, of reactor grade, will accumulate as a result of power reactor operation. Currently at least, the government is obligated to purchase this material. Quite obviously long-term uses for the material should be developed.

2. The recycling of plutonium yields a larger energy output from a given mass of raw fissionable material than would be achieved without recycling (recycle of U^{233} in a Th- U^{233} system would of course be competitive in this respect). It is stated that if plutonium is used in a reactor having a conversion ratio of 0.8, and is recycled six times, the energy available from 1 g of the initial plutonium equals that available from ~4 g of fuel consumed in a single-cycle system. The definition of a single-cycle system here is apparently a system which burns pure plutonium without conversion, and not the normal slightly enriched uranium system which is visualized for most reactors. In such reactors recycling of the U^{235} is generally anticipated, until the U^{235} is nearly all burned. In the process a significant amount of plutonium converted from U^{238} is inevitably burned. The factor of 4 is therefore something of an overestimate if one compares with the fuel systems currently visualized.

3. Recycled plutonium as an additive to natural uranium can take the place of slight enrichment

in certain reactors and thus allow these reactors, which otherwise would require fuel enriched by a diffusion plant, to operate with natural-uranium feed. The feasibility of a self-sustaining cycle of this kind depends on the enrichment required by the reactor and the conversion ratio. In the reference report are calculated curves connecting these two variables. As an example, one might consider a reactor which, if operating on slightly enriched uranium, would require enrichment of 1 per cent. According to the curves, such a reactor could be fed by natural uranium, with a self-sustaining plutonium recycle provided the conversion ratio is greater than 0.43. This figure holds if the fuel is enriched uniformly with plutonium. If the plutonium enrichment is added in the form of spikes, a conversion ratio at least as high as 0.6 is required.

4. In the same way that plutonium can be used to enrich natural uranium, it can also be used as an additive to depleted uranium from diffusion plants to bring its enrichment up to a point where it is usable as fuel in some reactors. Curves are also presented for this type of enrichment of 0.5 per cent U^{235} uranium feed. As an example, uniform enrichment with recycled plutonium will bring such uranium up to the equivalent of natural uranium, and will constitute a self-sustaining system, if the conversion ratio is as high as about 0.43. It is, of course, somewhat difficult to assess the value of this type of operation as a long-term proposition. The depleted uranium, even after the addition of the recycled plutonium, does not attain the equivalent of very high U^{235} enrichment and would have to be used in reactors capable of operation on natural or very slightly enriched material. Such reactors are quite feasible, but if they are to be used extensively, it may be that they would more suitably use natural-uranium fuel and thus reduce the demands on the diffusion plants.

The difficulties to be overcome in the use of recycled plutonium are stated to be the following:

1. It is necessary to develop suitable fuel elements of plutonium and/or plutonium-uranium mixtures.

2. The alpha activity and the resultant toxicity of plutonium pose technical problems in the handling of the plutonium and the fabrication of fuel elements. These problems will, of course, affect the cost of plutonium elements.

3. Because of the lower heat production per atom destroyed, and particularly because of the higher microscopic absorption cross section of plutonium, plutonium fuel elements will have a shorter residence time in the reactor than would uranium elements. This effect depends on the neutron temperature in the reactor since the plutonium cross section varies with temperature. At room temperature the throughput of plutonium fuel elements relative to elements of U^{235} having equal initial reactivity is about 1.4. At a temperature of 600°C the relative throughput is about 3. This characteristic means that fabrication costs must be kept low if the use of plutonium elements is to be economical.

In considering the actual worth of plutonium as a reactor fuel, the report referenced evaluates the material relative to U^{235} at \$17.11 per gram in terms of the energy output per gram. On this basis pure Pu^{239} should be worth \$14.10 per gram. The decrease relative to U^{235} is due to the circumstance that a greater fraction of the Pu^{239} is lost by neutron capture. The value of reactor grade plutonium, which would contain

only about 81 per cent fissionable isotope, would be \$11.42 per gram. Actually, in a competitive market the price of plutonium would very probably find a lower value relative to U^{235} because of its other disadvantages. Furthermore, it is hardly reasonable to base the economic value of plutonium on that of pure U^{235} since it is unlikely that many large power reactors will use pure U^{235} . It seems much more probable that plutonium, in the thermal reactor field, will eventually compete with slightly enriched uranium, for which the price lies in the range of 10 to 13 dollars per gram of U^{235} .

The reactor proposed for developmental work on plutonium recycle is a D_2O moderated, D_2O cooled reactor of 70 Mw heat output, which would generate steam in a heat exchanger, but not electricity. The reactor is of the pressure tube type, and the cool, unpressurized moderator is contained in a calandria tank. No discussion is given as to why this type of reactor is thought to be particularly suitable for the plutonium recycle program.

Reference

1. The Plutonium Recycle Program—A Résumé of the Concept, Program, and Facilities, HW-50700, June 12, 1957. (Unclassified AEC report.)

It is reported¹ that suspensions of fissionable material in NaK alloy are being investigated with the aim of developing a slurry that will be suitable as the fuel for a fast breeder reactor. Loop tests were made at high temperature on slurries of UO_2 in eutectic NaK. The UO_2 concentration was low in these tests (4.3 vol. %) whose purpose was to demonstrate the suspendability of the UO_2 at high temperatures and to develop techniques for studying the mechanical and physical properties of the suspensions. The UO_2 concentration used is considerably lower than that which would be suitable for a fast reactor, but it is stated to be one which would be suitable for an enriched thermal reactor.

The tests were carried out in two different high-temperature loops, constructed of $\frac{1}{2}$ -in. type 304 stainless-steel tubing. Circulation was provided by an electromagnetic pump. The circulating volumes in the two loops were 133 cm³ and ~160 cm³. The size distribution of the UO_2 particles was such that 61.6 per cent of the UO_2 was in the size range from 0 to 2 μ , 30.9 per cent was in the range 2 to 5 μ , and 2.7 per cent was in the range >10 μ . The measurements indicated that the particles were held in essentially complete suspension when the flow rate was 2.1 ft/sec or higher at 25°C and when the flow rate was 2.4 ft/sec or higher at 450°C. At a temperature of ~500°C, the particles came out of suspension. When the temperature was dropped to 480 to 490°C, the particles again went into suspension. This reversible effect was repeated many times over the course of the month's operation, and the temperature at which suspension failed was determined to be within ten degrees of the 500°C level. It was possible to decrease the extent of this dropping out of suspension by a treatment of the UO_2 which involved removal of some impurities and particularly the removal of oxygen contamination. The effect was eliminated entirely by mixing a small amount (~2.6 wt. %) of finely powdered uranium metal

with the UO_2 . It is believed that the effect of the uranium metal is to eliminate any traces of Na_2O in the fluid by reacting with the oxygen.

Some of the conclusions drawn from the tests are the following:

1. By the addition of a small amount of uranium metal to the system, the slurry is stabilized and may be circulated successfully at temperatures of 600°C.
 2. There is no evidence for corrosion or erosion of the stainless-steel tube (test durations were in the neighborhood of 400 hr).
 3. No change in the flow properties of the slurry was detected after circulating for 400 hr in the temperature range 400 to 600°C, although there was a shift toward larger diameters in the particle-size distribution below 3 μ .
 4. The slurry can be easily resuspended after prolonged settling.
 5. Unless the slurry is adversely affected by fission products, it appears that suspensions of UO_2 in NaK alloy can be potential reactor fuels.
- If further experiments establish the feasibility of the UO_2 -NaK slurry as a reactor fuel, it will become an additional candidate for use in high-temperature circulating fuel reactors. Its properties are quite different from those of the bismuth-uranium alloy and the fused-salt mixtures which are currently getting primary attention for such reactors, and therefore it would presumably add further design flexibility to the fluid metallic fuel concept. Tests in much larger loops, where the difficulties of avoiding contaminants are more nearly comparable to the situation in a reactor, would be of considerable interest.

Reference

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Compression Springs

Many safety-rod drives have incorporated in their design a spring whose function it is to initially accelerate the safety rod in order to cut down on the rod insertion time. The spring materials used are subjected to high irradiation fluxes in some cases which may change the spring characteristics. Irradiation tests were conducted by KAPL at Hanford on Haynes 25 and Inconel X springs.¹ The springs were irradiated in a compressed (stress = 80,000 psi) condition in water at 560 to 600°F and 2250 psi to integrated fast fluxes of 5.9×10^{18} nvt and 4.2×10^{19} nvt. Postirradiation examination showed no surface defects or cracks, and tests indicated very slight radiation damage. The spring constant of both materials increased slightly with a maximum change of 8 per cent for Haynes 25 and 5 per cent for Inconel X. Hardness did not exhibit the usual increase expected, but decreased slightly.

Ultrasonic Welding Machine

Continued effort by Du Pont on ultrasonic welding techniques resulted in the development of a continuous roller type ultrasonic welding machine capable of producing spiral-welded aluminum ribs at a maximum rate of 70 in./min and straight welds at two to four times that of spiral welds.² Improvements in the technique have substantially eliminated the cracking tendency for the aluminum alloy ribs.

Nondestructive Testing Apparatus

The application of commercial instruments to the nondestructive testing of Zircaloy-2 tub-

ing for PWR has been reported by Bettis Plant.³ A discussion is given in the report of the discontinuities detectable by the instrument such as cracks, seams, laps, pits, gouges, and wall thickness.

Evaporator for Processing

Radioactive Liquid Wastes

A de-entraining column has been developed which reduced the amount of nonvolatiles in the overheads to 1.5×10^{-7} times that of the evaporator bottoms.⁴ From bottom to top the evaporator consists of a dry bubble cap plate, a wet bubble cap plate, and a dry impingement cap plate. Authors say the column has low resistance to vapor flow, would require little maintenance, and is efficient over a wide range of vapor loads.

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PROGRESS ON SPECIFIC REACTOR TYPES

AQUEOUS HOMOGENEOUS REACTORS

Introduction

From time to time in this review of reactor development, the status of development of a specific reactor type will be reviewed in some detail. The objective is to establish a bench mark from which further progress can be measured and to define the developmental problems and their relations in sufficient detail to allow those who are not specialists in that reactor type to make some evaluation of future progress.

Table VIII-1 DESIGN PARAMETERS OF THE HOMOGENEOUS REACTOR TEST (HRT)

	Core	Blanket
Power, Mw (heat)	5	0.22
Pressure, lb/sq in.	2000	2000
Vessel	32 in. diameter 3/8 in. Zircaloy-2	60 in. diameter 4.4-in. stainless clad carbon steel
Volume, liters	290	1550
Specific power, kw/kg	1000	
Specific power, kw/liter	17	0.14
Conc. g U ²³⁵ /kg D ₂ O	9.6	0
Conc. CuSO ₄ , meters	0.005	0
Conc. H ₂ SO ₄ , meters	0.02	0
Circulation rate, gal/min	400	230
Inlet temperature, °C	256	278
Outlet temperature, °C	300	282
Volume gas generated, cu ft/S.T.P.	0.96	0.013

The name aqueous homogeneous reactors is derived from the fact that these reactors use solutions or slurries of the fuel in water (H₂O or D₂O) and the fuel is uniformly distributed throughout the aqueous moderator.

The first homogeneous reactors were the Los Alamos water boilers, LOPO, a zero power critical experiment, and HYPO, a 5-kw reactor which

first operated in December 1944. HYPO became SUPO when its power was increased to 45 kw in 1951. The homogeneous reactor project was started at Oak Ridge in 1950 to develop and build the Homogeneous Reactor Experiment (HRE), and to obtain information about the physics, chemistry, and engineering of aqueous systems operating under conditions that would produce useful power. The result was HRE-1, a small pilot plant, which, after about one year in operation, was dismantled in 1954.

Subsequently, work was begun on the construction of a larger reactor to demonstrate the use of the homogeneous reactor concept to produce power. In certain stages of its operation, the Homogeneous Reactor Test (HRT), also known as HRE-2, is expected to operate at 5 to 10 Mw. The core of this reactor is a uranyl sulfate solution, and the blanket region will be operated first with a D₂O reflector, then with uranyl sulfate solution, and finally, if possible, with a slurry of thorium oxide in D₂O. Design parameters¹⁻³ for the HRT are shown in Table VIII-1.

Originally the HRT was expected to be in operation in the latter part of 1956 or early part of 1957. Although the reactor construction was completed, preliminary nonnuclear tests at operating temperature and pressure revealed some stress corrosion cracking in the high pressure leak detector flanges and tubing, resulting from a high chloride ion contamination.³ This chloride contamination apparently resulted from the failure to remove all the die-drawing compound prior to annealing the tubes during manufacture. The trapped chloride was subsequently leached by water and caused the corrosion cracking in the leak detector pipe and flange system. Although there was doubt that the flanges would ever crack sufficiently to leak, it was decided to replace the faulty tubing and flanges rather than risk the possible replacement after the reactor had been operated at power and the system became radioactive. The HRT actually went critical on Dec. 27, 1957.

Following the successful operation of the HRT, the next step proposed in the power demonstration program is the construction of a 50-Mw breeder.

There has also been work on a number of aqueous homogeneous systems by groups other than the National Laboratories. Three of the more important efforts are listed in Table VIII-2. Phase 1 work⁴ on the Pennsylvania Advanced

is free of any neutron absorbing structural material and since the fluid state of the fuel allows continued removal of fission-product poisons. However, this high neutron economy is of value only if it is used in a productive manner in a fertile material such as U^{238} or thorium. The choice of the method of incorporating the fertile material is perhaps the most basic decision in the design of the reactor.

Table VIII-2 COMPARISON OF HOMOGENEOUS REACTOR CONCEPTS^{4,5}

	PAR*	Wolverine	Dutch
Type	Single-region slurry	Single-region solution	Single-region† slurry
Fuel	UO_3ThO_2 mixture	UO_2SO_4	UO_2
Moderator	D_2O	D_2O	H_2O
Power, Mw (heat)	550	27	0.250
Power, Mw (elec.)	150	10	
Core vessel	15 ft O.D. 6-in. steel $\frac{1}{4}$ -in. stainless steel	15 $\frac{3}{4}$ -in. sphere	12 $\frac{1}{2}$ -in. sphere
Pressure vessel	Same	Same	Same
Specific power, kw/kg		1600	
Specific power, kw/liter		14.6	
Outlet temperature		570	480
Outlet pressure		1900	750

* Has 7.2-Mw gas superheat in addition.

† Has BeO reflector.

Reactor (PAR) began in 1955 by Westinghouse and the Pennsylvania Power and Light Company of Allentown, Pa. It was announced at that time that construction of the plant was conditional upon demonstrating, by about January 1958, that the reactor would be technically feasible and economically competitive. The Wolverine plant⁵ (Wolverine Electric Cooperative, Hersey, Mich.) is scheduled to be built by 1959. The Dutch reactor is experimental and is being operated as a critical experiment before being built with power-removal equipment.⁵

Fundamental Characteristics

In the following paragraphs the fundamental characteristics which determine the design of an aqueous homogeneous reactor are discussed briefly.

Neutron Economy

The neutron economy of a homogeneous reactor is inherently quite good since the core region

Chemical Form of Fuel and Fertile Material

Fundamental to the choice of the method of incorporating fertile material are the characteristics of the possible fuel-moderator and fertile-material-moderator mixtures (i.e., slurries or solutions). Thorium solubility limitations (~70 g of sulfate per liter)⁶ and plutonium (3 mg/liter)² restrict the use of these materials in solution form. Except for corrosion considerations and the limited solubility of converted plutonium, there is no limit on the solubility of uranyl sulfate until a concentration well above that required in a reactor is reached. There are therefore three possible arrangements for a breeder or converter reactor: (1) low-concentration, high-enrichment solution core, with a blanket slurry of fertile material (thorium or U^{238}) in a two-region reactor; (2) the use of high-concentration, low-enrichment fuel in slurry form; or (3) the use of a high-concentration, low-enrichment solution core with continuous separation of insoluble produced plutonium. The latter two concepts may also use a blanket, but this would not

be a necessity for reasonable conversion ratios. In the case of a blanket type two-region reactor, the neutron economy will remain good only if a low absorption core vessel is used.

Moderators

For economic breeding or conversion of the fertile material, it is necessary to use heavy water as moderator and coolant. The use of light water in the core solution would require very high concentrations of U^{235} to reduce the relative parasitic absorption in hydrogen and would result in high costs from the large fuel inventory unless very high power densities can be achieved. If a low concentration of U^{235} were used, the high parasitic absorption would result in a low conversion ratio and hence high burn-up costs. For some purposes, such as research or nonbreeding burner reactors, ordinary water might be used as moderator.

Power Density

Theoretically, since no heat transfer is required within the reactor core region itself, the power density in the reactor may be quite high, limited only by fuel circulation rate and permissible temperature rise. Present designs, however, achieve rather modest power densities in the range of 15 to 20 kw/liter,^{2,3,5} primarily because of the limitations imposed by the increased corrosion rate under the influence of radiation. Since this corrosion rate is believed to be a function of the fission rate near the material in question (e.g., vessel walls), the attainment of a uniform power distribution is not necessarily a desirable goal in the homogeneous reactor, but rather, a power distribution which is peaked near the center of the vessel and low near the vessel walls may give best performance. High power density is desirable to minimize reactor vessel size for a given power output and to minimize inventory charges for D_2O and fuel. The latter charge depends, of course, on the size of the fuel inventory. In the homogeneous reactor the core fuel inventory may be low because of the low parasitic absorption. However, the total reactor inventory must include the remainder of the fuel in the circulating fluid system, the extent of which depends on heat-transfer rates in the heat exchangers as well as on other variables.

Fuel Reprocessing and Handling

Elimination of fuel fabrication costs is an important economic advantage of the homogeneous reactor. Fuel reprocessing for these reactors is usually thought of as a continuous or semi-continuous on-site process; however, reprocessing at a large central plant is also a possibility. The advantage of an on-site processing plant is the lower fuel inventory and freedom from the necessity for handling and shipping radioactive material in large batches. A possible disadvantage is the presumably higher unit processing cost associated with the operation of a small-scale plant.

Reactor Stability

The reactors possess a very high degree of nuclear stability.^{2,7-11} Gross changes in operation conditions can be achieved by adjusting the fuel concentration. The strong negative temperature coefficient of reactivity provides regulation and prevents harmful power excursions. Control rods and plates are usually considered to be unnecessary.

Operating Temperature

A characteristic common to all water cooled reactors is that high pressures are associated with the high temperatures required for the efficient production of power. For example, the vapor pressure of water is 1200 psi at 572°F (300°C) and 2400 psi at 662°F (350°C). In addition, circulating fuel reactors are operated with added pressure to prevent cavitation in the pumps and to minimize the volume of gases produced by decomposition of the moderator. Therefore the size and cost of the pressure vessel, which must withstand the pressure required, impose a limit on the temperature of the reactor. Additional limitations are imposed by the increased corrosion rate at higher temperatures and by the fact that uranyl sulfate solutions separate into two phases of different concentrations at temperatures above about 300°C (references 2 and 3).

Radiolytic Decomposition

Deuterium and oxygen are produced by the radiolytic decomposition of the moderator and must be recombined safely. The rate of gas pro-

duction is much higher in homogeneous reactors than in solid fuel reactors since the D_2O is exposed to the fission recoil fragments.

Problems of Radioactive Fluid Fuels

Large volumes of very highly radioactive materials are present in a fluid state at high temperature and high pressure. Provision must be made for containing those materials if they leak from the reactor vessels or piping. Pumps, piping, and heat exchangers become radioactive from contact with the fuel. Their replacement or repair can be difficult and expensive.

The use of fluid fuel results in a simple design of the reactor proper. However, the uranium, plutonium, and thorium compounds, water, and materials of construction constitute a very complex chemical system. In order to handle this system adequately, to cope with the problems of the high radioactivity, and to meet the demands of water recombination and fuel reprocessing, the total system may become quite complex.

Reactor Types

Homogeneous reactors may be classified according to several design parameters: (Table VIII-3 lists several reactor types for which some design studies have been completed.)

tors may be further subdivided into those where circulation is affected by natural convection or forced convection.

2. The pressure vessel may be a one- or two-region design. In the one-region concept the reactor is a sphere in which the heat generation and breeding occur. The two-region concept is usually two concentric spheres. In the inner sphere, or core, the majority of the power is developed, whereas in the outer region, or blanket, breeding and the remainder of the power generation occur.

The three main fluid loops in the reactor system are the core fluid, blanket fluid, and turbine steam. Power produced in the blanket may either be added to the core power or possibly used to superheat the turbine steam.

3. In general, the two types of core fuel presently under consideration are uranyl sulfate solution and slurries of uranium oxide. Although other soluble salts of uranium exist, such as the nitrate, phosphate, or fluoride, more is known about the uranyl sulfate solution. Under the proper conditions, uranyl sulfate may be contained in contact with stainless steel, zirconium, or titanium and with usable low corrosion rates. Uranyl sulfate solutions in D_2O separate into two phases² at temperatures of about 300°C; this characteristic may limit the temperature rise of the core solution.

Table VIII-3 DESIGN STUDIES FOR REACTOR TYPES

Name	Type	Core	Blanket	Power, Mw	Temp. and press.	Conv. ratio	Designer	Ref.
Thorium Breeder Power Station, TBR	Two region	UO_2SO_4 solution	ThO_2 slurry	300	436°F	1.11	ORNL	2
Homogeneous Natural Convection Power Reactor	Single region with diffuser	UO_2SO_4 solution, no chemical processing		10	482°F	Burner	ORSORT	12
Ultimate Homogeneous Reactor, UHR	Two region	UO_2SO_4 solution	ThO_2 slurry, boiling and used for superheat	200	600°F, 560 psi with superheat	1.12	ORSORT	13
Boiling Homogeneous Breeder Reactor	Single region	UO_2ThO_2 slurry		500	545°F, 1000 psi	1.03	ORSORT	14
Thorium Rod Breeder Reactor	Two region	UO_2SO_4 solution	Th rods in D_2O	480	535°F	1.10	ORSORT	15

1. Method of removing heat: whether the reactor is cooled by circulation of the fuel through external heat exchangers or whether the fuel is allowed to boil and heat is extracted by condensation of the vapor. Circulating fuel reac-

4. Fertile materials for homogeneous breeder or converter reactors may also be solutions, slurries, or solid rods. Soluble thorium compounds in sufficient concentrations for breeding have not proved feasible. The use of thorium ni-

trate would require low cross-section separated N^{15} which would impose an economic penalty.¹⁶ Recently the decomposition of thorium nitrate solutions by radiation has been investigated.¹⁷ High concentrations of uranyl sulfate for plutonium production may be economical for existing values of plutonium. A typical blanket for plutonium production would be a 1.4M UO_2SO_4 solution, circulated through a hydroclone to concentrate and separate the insoluble PuO_2 (reference 2). This system is proposed for the second stage of operation of the HRT reactor. One of the principal concerns is the adsorption of plutonium on the metal walls of the system, incorporated into the oxide corrosion layer. The only way to remove adsorbed plutonium is to destroy, periodically, the corrosion film.^{3,18} Solubility of plutonium in static conditions is somewhat unpredictable and apparently depends on a large number of factors, such as temperature, previous history, degree of disproportionation into $Pu(III)$ and $Pu(VI)$, and surface area of vessel walls.² No information is currently available of behavior of plutonium in dynamic systems at the reactor operating conditions.

The majority of effort so far has been directed toward the use of thorium or thorium oxide as the fertile material in order to take advantage of the high thermal η value of U^{233} . Most of the effort has been concerned with thorium oxide slurries; however, the use of thorium metal rods has been suggested.¹⁵ Although the use of thorium rods eliminates the problem of handling slurries, the cost of power would be higher, principally because of the higher chemical reprocessing costs and the lower attainable specific power in the blanket.

Thorium oxide, as usually prepared from the thermal decomposition of thorium oxalate, is a white granular solid (fluorite crystal structure) with a crystal density of 10.06 (X ray).³ By suitable control of the conditions under which the oxide is formed, the physical properties of the resulting oxide may be varied over a fairly wide range. However, little is known as to what physical and chemical properties are important in determining its handling characteristics as a slurry under reactor conditions.

Although the problems of corrosion and recombination appear to be under control in test loops, the problems of caking and settling still remain. There are some indications that the pH or sulfate concentration has a large effect on the caking and settling problem, although some ap-

parently identical systems show a different response. A considerable amount of development work remains before the thorium oxide system is ready for practical use.

Chemical Processing and Fuel Utilization

Because of the absence of fuel fabrication costs in the homogeneous reactor, and because of the latitude which is permissible in fuel loading of a given reactor, it is possible to maintain the reactivity over long periods of time by the simple addition of enriched fuel. It is therefore possible, at least in a D_2O moderated homogeneous reactor, to operate on enriched uranium in such a way that almost complete burn-up of the uranium is achieved without incurring large expenses for fuel refabrication and reprocessing. It may therefore be said that if enriched uranium is to be burned without producing converted fuel from fertile material the aqueous homogeneous reactor is one of the better ways of doing so. However, even with essentially complete burn-up of enriched uranium, the fuel burn-up cost is in the neighborhood of 3 mills/kw-hr, and, if really low fuel cost is to be achieved, the reactor must convert or breed new fissionable material. Such a process requires chemical processing of the fuel to remove fission products and to separate fissionable material from fertile material.

Most of the homogeneous reactors which are being considered seriously at the present time utilize thorium (in the form of oxide) as the fertile material. The Thorex process is a general process which can separate uranium, thorium, and fission products from one another. In many cases other simpler processes can be used to supplement the Thorex process. For example, gaseous fission products will normally bubble out of the fuel solution during reactor operation, and certain other fission products will form insoluble compounds in the fuel solution and can be removed by centrifugal separation. The actual combination of processes used will depend on the type of reactor under consideration.

The processes involved in processing a two-region reactor will be described in some detail. In this system, while the fuel is being consumed in the core, Pa^{233} , which decays to U^{233} , is produced in the blanket. The produced U^{233} must be removed from the blanket for re-use as fuel; any excess over that required for fuel replacement

will then represent the breeding gain. At the same time fission and corrosion products must be removed from the blanket and core.

In the reactor core fission products and corrosion products from the reactor vessel and piping accumulate. In order to maintain the highest possible conversion ratio, these poisons must be removed. Since a number of these products, such as the rare earths, strontium, barium, zirconium, and niobium, are insoluble,² it is highly desirable to remove them from the core solution. Furthermore, from a hazard viewpoint, it is desirable to maintain the concentration of fission products at a minimum level and to isolate them in a form such that they cannot be released to the surrounding area by a reactor accident.

Poisons encountered in the reactor core may be considered in three classes, i.e., soluble, insoluble, and gaseous products of both corrosion and fission. The rare gases, krypton and xenon, will be removed from the reactor along with the radiolytic deuterium and oxygen going to the recombination system.^{2,3} Iodine, which is one of the major poisons, may be removed by stripping with oxygen at reactor temperature and pressure, followed by backstripping into water.

Insoluble poisons may be removed from the core fluid by means of a hydroclone separator (hydrocyclone type of centrifugal separator). The concentration of soluble fission products may be kept low by continuous purging and replacement of a small volume of core fluid at a rate expressed in terms of the number of days required to process an entire core volume. The small volume thus purged is cooled and later combined with a portion of the blanket and sent to the chemical processing plant (Thorex) for recovery of the fissionable material.

Under the operating conditions, and in a uranyl sulfate solution, it is quite fortunate that the rare-earth sulfates, which contribute about 70 per cent of the fission-product poisoning effect, are relatively insoluble.² In addition the sulfates of iron, zirconium, and chromium hydrolyze to insoluble oxides. With the hydroclone separators^{2,3} operating at a concentration factor of 25, the insoluble poisons will be efficiently separated. In the HRT, for example, which is expected to produce about 1.36 g of insolubles per day, the residual concentration in the reactor will be 50 to 70 mg/liter (55 to 75 g total). The separated fraction, collected in the underflow receiver of the hydroclone, will be removed for processing once each week. Since this receiver will also

contain 2 gal of fuel solution, an entire core volume will be chemically processed once every 38 weeks. If the underflow receiver were removed daily, a full core volume would be processed in 38 days. Hence the cycle time determines the residual level of soluble poison in the equilibrium reactor.

In order to minimize radiation damage of the chemical reagents used in the processing stages, a certain amount of time must be allowed for decay of fission-product radioactivity. This cooling period depends on the initial level of radioactivity and the relative cost of replacing reagents as compared to the higher inventory charge incurred by a long cooling period.

In the Thorex process,² after the fuel and fertile material are dissolved in strong nitric acid, uranium and thorium are coextracted from the fission products and protoactinium into tributyl phosphate. In a second partitioning column thorium is preferentially stripped into a dilute nitric acid solution. The thorium nitrate product is then passed to oxalate precipitation and calcination operations for refabrication of blanket material. The tributyl phosphate leaving the thorium partitioning tower passes to a third tower where the uranium is stripped. Uranium is adsorbed from the resulting solution onto a cation-exchange resin and is subsequently eluted with a sulfuric acid solution to yield a uranyl sulfate solution to be returned to the reactor core as fuel.

An alternate method of solution core processing has recently been proposed.³ In this method the precipitates from the hydroclone separator are first mechanically separated, leaving the uranium and some soluble poisons in the supernatant liquid. The uranium is then recovered from solution by peroxide precipitation as the insoluble UO_4 , which may be readily converted to the sulfate by thermal decomposition in a dilute sulfuric acid solution. Although the product purity is not so high as in the complete Thorex process, separation factors of greater than 10 from dissolved poisons, with 99.9 per cent recovery of the uranium, have been demonstrated on a laboratory scale. Such a process may be more economical, especially in the smaller power plants, where it would be desirable to ship the irradiated fertile material (ThO_2) to a central processing plant but to process the fuel solution on site to minimize fuel inventory charges.

Cleanup of uranyl sulfate solutions, using hydroclones to remove corrosion and fission-

product precipitates, has been demonstrated in the HRT chemical plant.² Some uncertainty exists as to the manner that precipitates will originate and be distributed in an operating power reactor, and the operation of HRT is expected to answer this question.

In single-region reactors the combination of processes would be somewhat different. The straight burner reactor is generally conceived as a reactor without continuous processing other than the removal of gaseous fission products. Eventually, when it is no longer feasible to maintain reactivity by the addition of fuel, the reactor fluid would be removed, the uranium isotopes separated from the liquid and processed as a batch. The single-region thorium converter using a slurry of uranium and thorium oxide would be processed in a semicontinuous manner by the Thorex process.

Corrosion

Corrosion enters the aqueous homogeneous reactor technology not entirely as a feasibility problem but as a limiting factor in reactor performance as well, for it has been shown that corrosion rates increase as the fission power density increases. No methods have been found for reducing corrosion rates to the point where they

Table VIII-4 ZIRCALOY-2 CORROSION
IN IN-PILE TESTS²

Kw/liter	Corrosion rate, mills per year		
	280°C	250°C	250°C*
1	3		
4	9		
4.5	10	5.7	
9	13		
10		8	0.8
30	40		

*0.4M H₂SO₄ added.

are obviously negligible. Rather, it appears that in designing the homogeneous reactor one will strive for conditions which give a corrosion rate sufficiently low to be tolerable. The decision as to how high a corrosion rate can be tolerated will involve a good deal of judgment until enough reactors have been built and operated to give some background of experience. Obviously such

decisions involve a judgment as to whether experimentally determined corrosion rates can be relied on as representative of all parts of the reactor system. The experiments which have been made appear to indicate that corrosion rates over extended surfaces are relatively uniform and that excessive local rates or pitting are not to be expected.

The chloride stress corrosion which has caused delay in the HRT does not represent a corrosion problem which is characteristic or peculiar to the homogeneous reactor. It occurred in the leak detector system which was not exposed to the fuel solution.

Uranyl Sulfate Solution

The principal materials of interest for use in the core of a homogeneous reactor are zirconium, stainless steel (austenitic), and titanium. In the case of the uranyl sulfate core solution corrosion of zirconium and titanium, the sulfate salts formed are hydrolytically unstable and decompose to form a protective film of oxide. However, in the case of stainless-steel corrosion, the initial sulfate salts of iron (ferrous) and nickel are soluble and stable and hence would not form a protective coating. The presence of an oxidizing agent, such as oxygen gas, would oxidize the ferrous salt to the ferric state, which is hydrolytically unstable and would decompose to form a protective coating of oxide. Nickel sulfate, however, is not oxidized and would remain in solution. The nickel concentration in solution thus may be used as a measure of the degree of corrosion of stainless steel.

Out-of-pile tests^{2,3} have indicated that both Zircaloy-2 and titanium are essentially completely resistant to uranyl sulfate solutions. The addition of certain alloying elements, such as aluminum, chromium, niobium, cerium, silver, or yttrium, to pure zirconium increases the out-of-pile corrosion rate in uranyl sulfate solution.

Under in-pile conditions the corrosion rate of Zircaloy-2 is markedly increased and is a function of power density, temperature, and acidity. Typical results of in-pile test loops are shown in Table VIII-4.

The effect of power density and temperature has been found to be expressed reasonably well by an equation of the form³

$$CR = A (PD) (1 - e^{-B/CR^{1.5}})$$

where CR = the corrosion rate

PD = the power density

A = a constant which depends upon temperature and solution composition

B = a constant which is temperature dependent

Typical results are given in Table VIII-5. The first set of conditions represent fairly closely the operating condition of the HRT reactor. These values are for experiments with ordinary H₂O, and the effect of D₂O is not known.

Table VIII-5 EQUATION CONSTANTS FOR
CORROSION RATES OF ZIRCALOY-2
[CR = A (PD) (1 - e^{-B/CR^{1.5}})]

	A	B
0.04 UO ₂ SO ₄ , 0.008 CuSO ₄ H ₂ SO ₄ 0.02 (mean) T = 280°C	2.45	95
0.17 UO ₂ SO ₄ , 0.015 CuSO ₄ H ₂ SO ₄ 0.028 (mean) T = 280°C	1.04	95
0.17 UO ₂ SO ₄ , 0.03 CuSO ₄ 0.02 - 0.03 H ₂ SO ₄ T = 250°C	1.25	6.5

Some of the tests have indicated that the addition of H₂SO₄ or Li₂SO₄ reduces the corrosion of Zircaloy-2 by HRT fuel solution. However, insufficient data are available to answer this question with any degree of reliability.

Corrosion of titanium showed very much the same general trend as did Zircaloy-2. However, the corrosion of titanium was a factor of 3 or 4 less than that of Zircaloy-2 under the same conditions.

Corrosion of type 347 stainless steel, used for construction of a large portion of the system other than the core vessel, is subject to a number of interrelated factors. At low temperatures the character of 347 stainless steel is such that very low corrosion rates are observed up to a temperature of about 150°C. Above 150°C the corrosion rate increases markedly with temperature² (becomes 29 mils per year at 225°C). At higher temperatures the protective oxide film begins to form and the corrosion rate drops to a quite small value in static systems. However, under flow conditions, as the flow rate increases, a certain critical velocity is reached, above which the corrosion rate rather abruptly increases. The protective coating is apparently stripped at flow rates greater than the critical velocity, exposing the bare metal to the rapid attack of the fuel solution. In homogeneous reactor design it is very important to avoid areas of high turbu-

lence in order to avoid a localized high corrosion rate.

The critical velocity is affected by several conditions that may exist in the core or related piping. As the uranyl concentration is increased, the critical velocity decreases, and the bare-metal corrosion rate increases. As the temperature increases the critical velocity also increases, and the bare-metal corrosion rate increases. As acid is added to the system, the critical velocity is reduced. It does not appear feasible to operate with H₂SO₄ concentrations above about 0.05M, which is not sufficient to inhibit the corrosion of the Zircaloy-2.

It can generally be expected that corrosion rates will be lower when the reactor is operating at a temperature of about 250°C. Increases or decreases from this temperature will increase corrosion rate.

The addition of lithium sulfate in equimolar concentration greatly reduces the corrosion of stainless steel, and in addition the temperature of the phase separation of the uranyl solution is extended. However, the presence of Li₂SO₄ increases the hydrolytic precipitation of uranium at reactor temperature.^{16,19}

The addition of 0.04 BeSO₄ also appears to reduce the corrosion rate of stainless steel,¹⁶ although at higher temperatures some beryllium is lost from solution.

Studies of stress corrosion cracking of austenitic stainless steels have indicated that chloride or bromide ions are very consistent in producing cracks in a relatively short time. Tests of HRT fuel solution with as little as 25 ppm of chloride produced cracks in stainless-steel specimens (stressed to 15,000 psi) in less than 200 hr. Chloride ion concentrations of 10 ppm or less appear to be harmless.³

Corrosion-Slurries

The mechanism for corrosion in slurries is very nearly the same as that for solutions, i.e., chemical attack of the metal by the solution.^{2,3} Titanium, stainless steel, and Zircaloy are capable of providing a protective film. Wearing away of the protective film by the solid particles (ThO₂) in the slurry exposes the base metal to attack by the water. Chemically inert material such as gold and platinum show very little corrosion with slurries. Figure 4 shows the relative corrosion rates of several metals to aqueous slurries of ThO₂.

The corrosion rate of type 347 stainless steel may be minimized by controlling the particle size, the pH of the water, and by the addition of an oxygen gas overpressure.

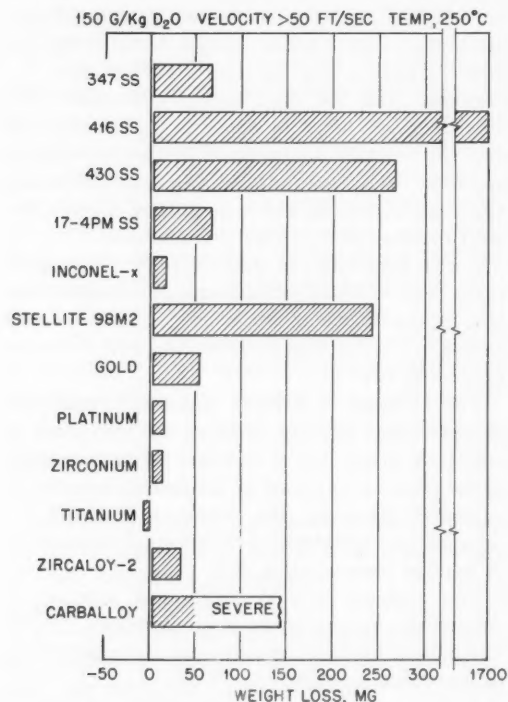


Figure 4—Relative attack by ThO_2 slurries on some materials.²

It has recently been reported^{4,16,20} that the corrosion-erosion rate by thorium oxide slurries, although fairly rapid at the beginning of the test, decreases markedly after the first 600 to 700 hr, presumably because the slurry particles become more nearly spherical as they are circulated. It has been reported⁴ that the designers of the PAR expect to be able to design so that the corrosion-erosion rate will be less than 1 mil/year in the major portions of the plant.

Control, Safety, and Stability

Aqueous homogeneous reactors rely for control on the variable fuel concentration and on the temperature coefficient of reactivity. Since the fission heat is produced directly in the fuel solution, the reactor temperature reaches a new equilibrium value quickly after any change in

reactivity or in heat removal rate, the equilibrium value being that at which the rate of heat production is equal to the rate of removal. The reactor is therefore self-regulating with respect to power. In terms of steady-state operation, the fuel concentration does not determine the operating power, but rather the operating temperature. The reactor automatically matches its power to the load demand, without the action of control rods. For example, an increase in heat removal would result in cooler fuel solution being pumped into the core, which, as a consequence of the very large negative temperature coefficient of reactivity ($-0.002 \Delta k/^\circ\text{C}$ for the HRT), would result in an increase in reactivity and an increase in power.^{7,8} The power would level out at the new demand level, and the fuel solution temperature would stabilize at some level which would make the reactor just critical. This temperature level might or might not be the same as the original level, depending on whether reactivity changes were associated with the power increase.

It is anticipated that in a typical case the reactor will be started by simultaneously charging the blanket slurry (if a two-region reactor as in the HRT) into the blanket region and D_2O into the core volume. When both the core and blanket systems are filled, they will be pressurized and the circulating pumps will be started. If desirable, the circulating fluids may be heated prior to beginning the injection of concentrated fuel. When the critical concentration is reached, the chain reaction will begin and the reactor temperature will rise with continued addition of fuel. As the temperature approaches the desired operating temperature, steam will be extracted to start the turbines. Final adjustment of the fuel concentration will be made with the system operating at rated output.

The process will be reversed in a normal shutdown of the reactor. Fuel from the core will be pumped to the storage tanks, where the D_2O will be distilled and returned to the core to replace the fuel, and the temperature will then be slowly reduced until the reactor is cool. In an emergency the core solution may be dumped to the storage tanks in approximately 10 min.²

The safety of the HRT reactor has been studied under a variety of operating conditions and with various blanket materials.⁸ The studies treat the effects of rapid addition of reactivity to the reactor. Such additions cause rapid power increases which are terminated when the fuel solution

reaches a temperature high enough to remove the excess reactivity. The rapid expansion of the fuel solution during heating exerts a pressure on the reactor walls; this pressure is considered the limiting factor in determining reactor safety. To eliminate operational hazards, a conservative value of 400 psi was considered the maximum permissible pressure rise which would not endanger the core vessel, whereas the maximum safe pressure of the main pressure vessel was considered to be 4000 psi, both values being less than the allowable yield stresses by ASME standards. The rates of reactivity additions, found in this study to be the maximum allowable for the design safety criteria, are summarized in Table VIII-6 and are based on an initial power level

A number of imaginable operational accidents were also investigated. These included addition of cold fuel solution into the hot critical core by misoperation of the heat exchangers and/or pumps; addition of concentrated fuel solution into the critical reactor; possibility of reaching criticality before the core is filled; and radiation induced or otherwise accidental rupture of the zirconium core vessel. In all cases the rate of reactivity addition appeared to be within safe limits, assuming the effects of radiation on the metallurgical character of the primary pressure vessel to be negligible.

A number of experimental investigations of the safety of aqueous homogeneous reactors have been made and more are being planned.^{10,11} These

Table VIII-6 SUMMARY OF HRT SAFETY CALCULATIONS⁸

Conditions	Blanket material			
	D ₂ O	335 g U/kg D ₂ O	633 g Th/kg D ₂ O	1349 g Th/kg D ₂ O
Rate of reactivity addition required to increase core pressure by 400 psi, % Δk /sec	0.8	0.84	0.80	0.75
Instantaneous reactivity addition required to increase core pressure by 400 psi, % Δk	1.9	1.6	1.3	1.1
Rate of reactivity addition required to increase reactor pressure by 4000 psi, % Δk /sec	3.1	3.0	2.8	2.6
Instantaneous reactivity addition required to increase reactor pressure by 4000 psi, % Δk	3.5	2.8	2.0	1.6

corresponding to only the source power. At the reactor power level the allowable rates of reactivity additions would be considerably greater.

The addition of excess reactivity would result in an increase in temperature of the fluid. Since this fluid occupies a large fraction of the available volume, and is essentially incompressible, the thermal expansion of the fluid results in a pressure increase. The temperature increase, of itself, is of no danger, and the corresponding pressure rise then becomes the determining factor for safety considerations.

The physical operations which are permitted are based on the reactivity additions which the associated physical event would introduce into the reactor. In all cases physical operations are restricted so that the rate of reactivity addition is limited to a maximum value of 0.8 per cent Δk /sec.

experiments have confirmed the self-regulating feature of such reactors and serve as bases for safety calculations.²¹

It can probably be said with some certainty that the greatest potential source of explosive energy release in the aqueous homogeneous reactor is the thermal energy of the high pressure fluid in the primary reactor system and that no large additional energy release is to be expected from nuclear sources. High pressures generated by the rapid production of nuclear energy during a reactivity accident could, however, conceivably cause a failure of the primary system. Even if this possibility did not exist, the possibility of a major rupture of the system could not be eliminated, particularly in view of the uncertainties which exist in the corrosion picture. It would therefore seem that the containment requirements on the housing for an

aqueous homogeneous plant would be a very high degree of leaktightness, the ability to withstand pressures resulting from the sudden liberation of the hot fuel solution, and adequate protection from any missiles which might conceivably result from a rupture of the pressure vessel or any other part of the primary system.

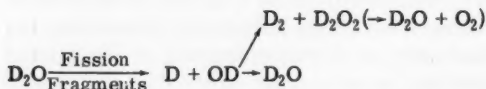
The stability analysis of the HRT has been based on the criteria that operating conditions must be such that a system disturbance does not build up in amplitude with time. In general, instability problems would most likely be expected to occur in small-size high power density reactors. The stability problem^{8,9} may be divided into three groups: (1) nuclear, (2) load demand, and (3) "walkaway" or physical stability. The nuclear stability studies were concerned with high-frequency nuclear power oscillations. Results of this study indicate that the fuel fluid circulation tends to damp out this type of oscillation for the operating conditions of the HRT.

Load demand variations will be of a time interval long compared to the nuclear power oscillations and are controlled largely by the temperature coefficient of reactivity. For the HRT the system was found to be stable for all variations in load demand.

Physical stability, or "walkaway" refers to the long-range changes occurring from radiolytic decomposition of the D₂O and slow changes in the composition of the fuel solution. This is of a sufficient time duration (of the order of minutes) that it can be controlled.

Hydrolytic Gas Production and Recombination

In a homogeneous reactor the fuel solvent (D₂O) is directly exposed to the highly energetic high mass fission fragments which produce rather extensive decomposition of the heavy water. The primary products of the fission fragments are the free radicals, D and OD. These may react to produce deuterium gas, deuterium peroxide, and recombined water.²



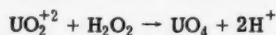
The yield of the radiolytic decomposition of D₂O is essentially constant with temperature and

amounts to ~10 cu ft (STP) of gas per minute per megawatt of reactor power.² It has been suggested that the deuterium and oxygen gas formed could be recombined in some kind of external flame recombiner¹³ and that the heat from the reaction could be used for superheat and reheat. This would require some sort of inert gas or steam in a pressure letdown system, to remove the deuterium and oxygen from the high pressure system and to prevent the accumulation of explosive concentrations. For a power reactor of the proposed type, operating at 450 Mw (heat), the energy available from the radiolytic gas recombination (4500 cu ft/min) would amount to about 10⁶ Btu/min or 17 Mw.

There are two possible hazards resulting from the large volume of gas formation. First, a sudden collapse of the gas bubbles in the reactor could result in the addition of large amounts of reactivity. Second, the possible accumulation of explosive mixtures of deuterium-oxygen gas in the reactor system could lead to damage of the reactor vessels.

Between 1950 and 1953 the ability of dissolved cupric salts to catalyze the homogeneous recombination of hydrogen and oxygen was discovered and developed. These gases have been found to recombine smoothly and rapidly at operating temperatures when catalyzed by dissolved copper sulfate. This reaction is a first-order reaction depending on the concentrations of hydrogen and copper in solution. By control of the ratio of copper concentration to uranyl concentration, it is possible to recombine any chosen fraction of the radiolytic gases.² Some gas production appears desirable to sweep out gaseous fission-product poisons. Examination of a large number of inorganic compounds has not revealed any homogeneous catalyst superior to the copper ion.

The second product of the heavy ion bombardment of water is peroxide formation. By the reaction



the partially soluble uranium peroxide will be formed and may be precipitated if the concentration becomes sufficiently high. The rate of peroxide decomposition is strongly temperature dependent and, in addition, is catalyzed by the presence of a number of impurities, such as iron. At the operating temperature of a homogeneous reactor, and in the presence of corrosion

and fission products, the problem of peroxide precipitation of uranium is not expected to exist.²

In a thorium oxide blanket the problem of radiolytic decomposition of the aqueous phase will also exist.³ Although the ThO_2 itself exhibits some catalytic activity, its effect is not sufficient to recombine the deuterium and oxygen. Copper ion catalyst, which was effective in the core solution, is chemically unstable under the conditions existing in the blanket, and the precipitated copper oxide is ineffective as a catalyst. Several materials, however, have been found which are quite effective in catalyzing the recombination reaction. Palladium, added as oxide, in concentration of 0.006M, and reduced to the metal, yielded high reaction rates at low temperatures and unmeasurably fast recombination rates above 120°C. High catalytic activity has also been observed for MoO_3 in the concentration region of 0.05M. Both of these catalysts are effective in essentially completely catalyzing the recombination during reactor irradiation of aqueous thorium slurries at the approximate average power densities expected in large-scale thorium breeder reactors.

Components

The basic simplicity of the homogeneous reactor concept has been emphasized by referring to it as a "a pot, a pump, and a pipe." Although this description does not do justice to the maze of plumbing which goes into the reactor installation, it does define the major components. To provide such components for the HRT, it was necessary to develop a number of specialized components and fabrication techniques.² The core vessel of the HRT reactor is a 32-in.-diameter sphere, constructed by shaping and welding 13 different pieces of Zircaloy-2 metal, with diffuser screens mounted on the interior. This vessel has been successfully constructed and tested.

Surrounding the core vessel is a stainless-steel-clad carbon steel pressure vessel $4\frac{1}{2}$ in. thick; outside the pressure vessel is the outer blast shield of $1\frac{1}{2}$ in. stainless steel, covered with cooling coils. The pressure vessel was formed into two hemispherical heads and was welded around the inside core vessel. Considerable development work was required to develop the girth weld of the pressure vessel since it could be made from the outside only and required

a transition from the stainless-steel cladding to the bulk carbon steel.

The canned rotor type pumps required for the reactor were built by the Westinghouse Electric Corporation. Initially, these pumps evidenced considerable corrosion and leakage, which required welding of the thermal barrier to the stator flange and the use of a titanium impeller. Such a pump has been operated with negligible corrosion in test for about 8600 hr at 255°C with the uranium fuel solution.

The large "pulsafeeder" fuel injection pumps were copies from the type developed for the HRE experimental reactor, with the later addition of a fine-mesh screen to prevent accumulation of small particles of trash. This pump has been the principal source of difficulty in the preliminary testing of the HRT system, primarily from corrosion and leakage at the Stellite balls and seats of the check valves. Some evidence indicates that these pumps may operate more satisfactorily with the fuel solution than with the pure water used for testing. However, they have been located in such a manner that replacement and repair will be facilitated.

The main heat exchangers, of which there are two, pass the fuel solution on the tube side and generate secondary steam on the shell side. Each exchanger contains 250 tubes, which are U-shaped, with $\frac{3}{8}$ -in. outside diameter and 0.065-in. wall thickness. The tubes are welded into the tube sheet.

The entire low-pressure system, including the concentration-dilution system, dump tanks, evaporators, entrainment separators, catalytic recombiner, and cold traps, was fabricated on a single frame and lowered into its cell. This unit has been tested and found to operate successfully on start-up, shutdown, and dump operation and during operation of the recombiner system at its maximum design rate of 15 cu ft of gas per minute.

Maintenance

Maintenance has long been recognized as one of the major problems associated with homogeneous reactors because of the large amount of highly radioactive fluid circulated through the system. The combination of unknown service life of components, serious consequences resulting from the release of radioactivity in the event of a failure, and the extreme difficulty of "hot"

maintenance of components has greatly influenced the design and layout of the homogeneous reactor concepts. The design of each component and its physical location must include careful consideration of the maintenance requirements. Experience has not been sufficient, as yet, to decide whether the philosophy of preventative maintenance, or that of operation until a failure occurs, will be more economical. No maintenance work can be performed in the nuclear part of the plant while it is operating because of the high radiation level, and it is necessary to duplicate a number of components in parallel to permit continued operation until maintenance can be scheduled.

Maintenance difficulties can increase the power cost in several ways. They may, of course, add to the operating cost and reduce the plant availability factor. On the other hand, if great effort is expended in the design and construction of the plant to provide means for remote maintenance, the availability factor may be kept high at the expense of an increase in capital cost of the plant. It can probably be predicted with some assurance that in the long run the economic pressure will be toward a high availability factor and that any inherent maintenance difficulties, which the homogeneous reactor has, will show up primarily in terms of capital cost and perhaps to some extent in terms of personnel requirements and operating cost.

Present maintenance philosophy involves the use of a number of cells containing the various components.^{2,3} These cells would serve as containment vessels in the event of a leak or rupture to prevent the spread of contamination. The same cells may be flooded with water to serve as shielding during maintenance operations. In general, maintenance work on the primary system will be performed remotely under some 15 or 20 ft of water and will require the design and development of specific remotely operated tools and jigs. Those fission and corrosion products which are water-borne must be prevented from diffusing through the shielding water and contaminating the surrounding area. Decontamination of the cells and their contained components by water and/or acid wash will reduce the hazard of contamination and exposure, but will not likely allow direct maintenance, particularly since delayed neutron activation will leave the components themselves radioactive. It is probable that it would be necessary to perform maintenance operations on some of the more radioactive

components, by means of remotely operated equipment, through seals that prevent leakage of gas or fluids from the cells.

In most cases it will be necessary to remove and replace a defective component rather than to attempt repair *in situ*. Repairs, if possible, may then be made in a remote location, after suitable decontamination and cooling operations. Such a program will require a sizable stock of spare parts and hence will add to the operating cost of the power plant.

To test the proposed operating scheme on a small scale, the HRT has been designed in such a manner that the various components are mounted in shield tanks which may be flooded with water for shielding. Special long-handled tools will permit workers to remove or perform routine maintenance on equipment inside the flooded pit. When it becomes necessary to open the system piping to remove equipment, the reactor fuel will be drained and flushed to outer storage tanks. The pipelines will then be flushed with distilled D_2O , and ice plugs will be frozen into the lines adjacent to the disconnect points. The particular piece of equipment may then be removed with the special tools designed for the purpose. To return to operating conditions, the procedure is reversed.

On three occasions 17 different dry-run remote handling jobs were successfully completed on the HRT system. Typical remote maintenance times³ are shown in Table VIII-7. For these jobs

Table VIII-7 TYPICAL REMOTE MAINTENANCE TIME REQUIREMENTS³

Item	Repair or replacement time
Circulating pumps	24 hr
Feed pumps	6
High-pressure valves	10
Low-pressure valves	6
Pressurizer heaters	2 hr/heater
Space coolers	4

about 24 hr additional time would be required to open the system, flood with water, and then to drain the cell and reseal the shield after the repairs were completed.

Remote maintenance practices similar to those developed for the reactor system will be employed in the chemical processing plant.

Initially, the annual maintenance cost contribution to the total cost of power produced will

probably be quite high. As experience and technology of homogeneous reactors develops, it is generally hoped that maintenance costs can be sizably decreased. The maintenance of homogeneous reactor systems appears mechanically feasible if research can provide answers to such problems as satisfactory decontamination methods, workable remote equipment capable of handling high-pressure systems, grinding and refinishing surfaces, and underwater welding techniques.

For the PAR a different maintenance principle has been proposed.⁴ Since the PAR uses an all-welded primary loop, and since strength-welding could be accomplished in air, and probably not underwater, the concept of underwater maintenance was abandoned. It is proposed to handle all repairs and replacements by the use of completely remote tools, on both high- and low-pressure systems. All four primary coolant loops will be identical so that any component can be used as a replacement in any loop and special maintenance tools may be used interchangeably. Auxiliary equipment will be segregated in separate cells, according to the expected level of radioactivity. In so far as possible, shielded windows permit observation viewing from access corridors within the biological shield. A 100-ton shielded crane will be the major maintenance tool and will also provide a facility for moving temporary lead shields. A number of techniques, such as remote strength-welding and testing, design of special manipulators, and leak detection apparatus, are currently being developed and tested.

Costs

The principal cost advantage of an aqueous homogeneous reactor lies in the relatively low parasitic absorption, which results in higher conversion ratio. It is expected that conversion ratios of 1.0 or greater will be attainable with either one- or two-region homogeneous reactors. Under these conditions the major power cost contributions will be from the fixed charges, i.e., plant amortization and operating expenses.

The fuel costs associated with electrical power from homogeneous reactors will be a function of feed cost, inventory charge, fuel processing cost, and operating conditions.

Most of the fuel cost estimates for homogeneous reactors have been based on an equilibrium

U^{233} - U^{235} -thorium fuel and on a large three-reactor power station having a total output of 375 Mw (elec.).^{20,22} With this type of reactor, operating with positive breeding gain (amount conversion ratio exceeds 1.0), the cost of fissionable feed material will be negative. The breeding gain, and hence the contribution of fissionable feed material cost, will depend on the relative blanket thickness and the allowed operating poison level. For a typical reactor the optimum blanket thickness, at 1500 g/liter, has been calculated as 2 ft for a 6-ft core and $2\frac{1}{4}$ ft for a 5-ft-diameter core. Optimum thorium concentration generally fell within the range of 1000 to 2000 g/liter; however, fuel costs obtained with blanket thorium concentration of 750 g/liter were only slightly larger. The higher thorium loadings would be required to maintain the breeding gain if thin blankets or small diameter cores were used.

Annual make-up of D_2O and thorium would be required. Cost of thorium make-up was generally assumed to be negligible (0.01 per cent or less), and ~5 per cent of the D_2O inventory was assumed as the annual make-up requirement. The effect of the build-up of radioactive Th^{228} from the decay of U^{232} was not taken into account and could affect the long-range cost contribution for thorium replacement or increased handling problems.

The poison level (defined as the ratio of the total macroscopic cross section per unit volume to the total fission cross section of the U^{233}) affects fuel cost, both by its effect on the breeding gain and on the inventory of fissionable material tied up in the reprocessing stages. The gross breeding ratio (neglecting processing losses) increases almost linearly with reduction in poison fraction level. This gain is counterbalanced by higher cost due to the increased chemical processing rate. The optimum values, from a fuel-cost standpoint, of the poison fraction was found to be 0.08 for slurry core systems and 0.07 for solution core reactors.

Based on several years of successful operation of a 200 kg/day Thorex pilot plant, the fixed charge (amortization and operating cost) of a processing plant capable of handling the proposed power station (375 Mw) has been estimated at \$5500 per day.^{20,22} For such a power station, operating at 80 per cent load factor, the net contribution to the power cost would be 0.764 mills/kw-hr. Since the total fixed processing costs were assumed to be independent of through-

put, the contribution to the fuel cost would be inversely proportional to the reactor power. Hence for a power station of 250 Mw, the contribution would be 1.19 mills/kw-hr, and for a 600-Mw reactor the contribution would be 0.48 mills/kw-hr.

In addition to the fixed cost, there is an operating charge which depends on the amount of material processed. This charge has been estimated at 50 cents per gram of uranium, \$3.00 per kilogram of thorium, and 35 cents per liter for heavy water recovery. It is possible that a newly proposed method of reprocessing core solution by peroxide precipitation may reduce both the reprocessing charge as well as the required inventory of U^{233} .

balanced by lower plant investment and other fixed charges in the single-region reactor because of its simpler construction and operation.

Until a large aqueous homogeneous plant has been at least designed in detail, estimates of power costs cannot be made with much confidence. Indeed, there are a number of questions, such as the dependence of corrosion on power density, which can have an important bearing on power costs but which are still in the research and development stage. It is difficult even to compare the fuel cost for the aqueous homogeneous reactor with that for a solid-fuel reactor because the homogeneous plant employs on-site chemical reprocessing, whereas the solid-fuel reactors generally use centralized plant reproc-

Table VIII-8 RELATIVE FUEL COSTS OF SOME TYPICAL REACTORS²⁰

125 Mw (Electrical)					
	A	B	C	D	E
Core diam., ft	6	5	4	6	14
Blanket thickness, ft	2	2	2½	2	Single region
Breeding gain	0.102	0.081	0.089	0.045	0.012
Core Th conc., g/liter	200	100	0	0	250
Blanket Th conc., g/liter	1000	1000	1000	1000	0
Est. cost, mills/kw-hr					
U inventory	0.27	0.20	0.15	0.11	0.38
D ₂ O inventory and loss	0.27	0.25	0.25	0.29	0.45
Th inventory and fuel	0.01	0.01	0.01	0.01	0.01
Fixed chem. processing	0.76	0.76	0.76	0.76	0.76
Operating core processing	0.13	0.13	0.07	0.08	0.19
Operating blanket processing	0.09	0.12	0.18	0.18	
Breeding credit	0.33	0.26	0.29	0.15	0.04
Net fuel cost	1.22	1.22	1.16	1.29	1.76

In Table VIII-8 representative costs of several reactor systems are given. These costs were calculated on the basis shown, and for a large 375-Mw station, with an integral processing plant.

There appears to be no decisive difference in fuel costs between solution-core and slurry-core systems. For small core diameters the solution core shows a slight advantage (gained from the use of hydroclone separation), but the power density (and hence corrosion problem) would be high. For larger core reactors the slurry core appears to have a slight advantage due to the larger breeding gain.

In general, the two-region reactor shows somewhat lower fuel costs (by about 0.5 mills/kw-hr) than does the single-region reactor. However, this advantage in fuel costs may be

essing. It can certainly be said that the homogeneous reactor has good potentialities for low fuel cost because of the good neutron economy and the absence of fabrication costs. If at any given time the fuel cost proves to be high because of the expense of on-site reprocessing, it can still be said that the cost reflects the technological development of that particular time, and is not an inherent characteristic of the system.

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Shim Control by Spectral Shift

In any reactor having a conversion ratio appreciably lower than unity, the reactivity will decrease with time. One of the major problems in the development of reactors with very long fuel lifetime is the maintenance of reactivity. If the construction of the reactor is such that frequent replacements of small fractions of the fuel can be made, the problem of reactivity change, at least after the reactor has reached equilibrium operation, is not a severe one. In pressurized-water reactors which are loaded and unloaded through large closures, the frequent replacement of fuel may be undesirable, and, even if frequent changes can be tolerated, there are practical limitations on the degree to which an equilibrium loading which is uniformly graded in irradiation can be achieved. A different approach in such reactors is to reload the complete core at one time and to maintain reactivity between reloadings by some kind of control element. The use of absorption type control rods for this purpose is not very attractive since the loss of neutrons to parasitic absorption in the rods decreases the conversion ratio still further and thus accentuates the loss of reactivity in the fuel.

A method of reactor control aimed at reducing the nonproductive consumption of neutrons has been suggested.^{1,2} Control of reactivity would be accomplished by varying the moderator slowing-down power and, hence, the core neutron spectrum during the operation of the reactor. The slowing-down power would be made small during the early life of the reactor, and resonance absorption in the fertile material would thereby be accentuated. In effect, this results in the fertile material acting as absorbing control rods, but the neutrons absorbed are used productively for the conversion of new fissionable material. A discussion of the difference between control by wastage of neutrons and control by productive use in conversion was given some time ago.³

The variation of slowing-down power would be accomplished by using a high concentration of D_2O as moderator at the start of core life and maintaining criticality through reactor lifetime by gradually diluting the moderator with light

water. In this manner the core initially could handle a large loading because of the high resonance capture in a D_2O moderated system of high fuel density. The result would be a much longer lifetime with less power distortion than in a comparable rod-controlled reactor and a higher conversion ratio due to the absorption of neutrons in fertile material rather than in a poison.

Calculations indicate that the limit of lifetime set by criticality could be extended in a Pressurized Water Reactor (PWR) type core by a factor of four or five over a rod-controlled system if a moderator of ~90 to 95 per cent D_2O were used initially and were gradually diluted through core lifetime to a concentration of ~40 to 50 per cent D_2O .

Since, at the present time, fuel lifetime may be determined by radiation damage effects rather than by criticality, it may be that the usefulness of this concept will not be realized in the immediate future. Among the problems of the system are the safety problem involved in any reactor system using a variable mixture of D_2O and light water as a moderator, because of the great differences in moderating power, and the emergency cooling problem, since there appears to be no possibility of using light water for emergency cooling.

The use of D_2O which is progressively contaminated by H_2O during the life of the fuel charge raises the question of the cost of re-enriching the D_2O . This cost may make a relatively small contribution to the total fuel cost if the reactor in question has high power density and does achieve very long fuel lifetime. It might be expected in such a reactor that the specific power relative to D_2O might be something like 25 kw/liter and that the specific power with respect to fuel might be something like 25 kw/kg. If it is assumed that the fuel enrichment would be approximately 3 per cent, then the cost of a kilogram of fuel would be \$375.00. This is considerably larger than the cost of a liter of D_2O (\$62.00). If the cost of re-enriching D_2O from, say, 50 to 90 per cent is small compared to the cost of pure D_2O (the cost of enriching U^{235} from 50 to 90 per cent is only 3 per cent of the cost of

enriching from the natural ratio to 90 per cent), then the re-enrichment cost for D_2O may indeed be small compared to the fuel burn-up cost.

Thermal Design Criteria for Pressurized Water Reactors

Westinghouse (Bettis) is pursuing a program designed to fill some of the gaps in the important but inadequately understood subjects of two-phase pressure drop and burn-out heat fluxes in pressurized water reactors.⁴ Better understanding of these topics should lead to uprating in power for reactors of this type since present design criteria are quite conservative.

It is generally believed that the power output from a pressurized water reactor may be increased substantially if local boiling is permitted during steady-state operation. Determining the extent to which this type operation reduces the margin of safety for both steady-state and transient conditions is the crucial problem which the Bettis program is attempting to resolve.

Data are now available for water at 2000 psi which indicate that the local boiling friction

factor is as much as 1.6 times as great as the isothermal friction factor, depending on the degree of boiling. New burn-out data have also been obtained which attempt to take into account the L/D effect for a heated channel.

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BORAX-IV

It is well known that boiling water reactors are characterized by large negative power coefficients of reactivity. This characteristic tends to limit the power level which can be attained for a given reactivity increase and therefore gives such reactors a degree of inherent safety. It also causes the power to be self-regulating under normal conditions of operation, but, like any inverse feedback characteristic, it introduces also the possibility of instability. In reactors I, II, and III of the BORAX series, such instabilities had been observed when the amount of reactivity compensated by steam reached a sufficiently high level. The instability took the form of oscillations whose amplitude increased rapidly until the peak powers reached were many times the normal operating power of the reactor. The characteristic frequency of these oscillations was between 1 and 2 cycles/sec.

When boiling reactors employing fuel elements having thick sections of uranium or oxides of uranium and/or thorium were considered, there was concern that the relatively long thermal time constants of such elements (which usually lie in the range $\frac{1}{2}$ to 1 sec) might accentuate the tendency toward instability. This concern was heightened by the recognition that the inherent power limiting capabilities of such poorly conducted fuel elements would probably be considerably less effective than those observed in the early BORAX reactors which employed thin fuel elements of aluminum-clad aluminum-uranium alloy. The BORAX-IV reactor was constructed to investigate the stability characteristics and self-limitation of power in a boiling reactor employing fuel elements of relatively long thermal time constant.

The BORAX-IV reactor has been operated¹ under transient and steady-state conditions. The reactor employs fuel elements containing $\text{UO}_2\text{-ThO}_2$ pellets in aluminum jackets with a lead filler.

The transient behavior tests were run with periods of 1 sec to 87 msec. The moderator temperature was below the boiling point at the time of peak power; the highest peak power

reached was 13.5 Mw. A shutdown mechanism appears to exist which terminates the power rise for these relatively mild excursions before boiling occurs in the moderator. The estimated effects of temperature change, formation of radiolytic gas, and Doppler coefficient account for only about one-third of the reactivity loss in such excursions. The other two-thirds of the reactivity loss has not been satisfactorily explained.

In steady-state boiling at atmospheric pressure, power densities as high as 30 kw/liter of coolant were obtained with $\Delta k/k$ of steam of ~ 1.2 per cent. The highest power attained was 4.6 Mw with 1.54 per cent $\Delta k/k$ in steam. Stability investigations based on the frequency response at various power levels indicated stability up to 4.7 Mw.

Although the steady boiling tests that have so far been made are only preliminary, the indications are that the stability of the oxide fueled boiling reactor is at least as good as that of the reactors employing highly conducting fuel plates, and probably a good deal better. On the basis of Experimental Boiling Water Reactor (EBWR) experience, it is predicted that the improved stability characteristic of the sluggish fuel plates will be more pronounced at higher operating pressures.

Preliminary results of oscillator experiments, whose purpose is to give more detailed information on the dynamic characteristics of the boiling water reactor, were presented. If such experiments can be used to predict the power level at which instability will occur in specific boiling reactors, they will be of great value, because one of the problems of the boiling reactor is that of limiting its operation to the stable region without penalizing its performance unduly. The difficulty in applying the oscillator technique for this purpose results from the background of irregular power variations which is characteristic of the boiling reactor.

Argonne Boiling Water Reactor

Reference 2 gives the preliminary design of the Argonne Boiling Water Reactor (ARBOR) fa-

cility, which was originally described in reference 3.

ARBOR is to be a highly flexible boiling water reactor experimental facility. The facility is designed for testing boiling water reactors with cores up to 6 ft in diameter and height, at any operating pressure between 600 and 2000 psig. Forced-circulation flows up to 36,000 gal/min are to be available, and heat removal capacities of 200 Mw in forced-circulation water coolers (at 600 psig system pressure) and 150 Mw in steam condensers (at 300 psig condensing pressure) are provided in the design. Higher rates of heat removal will be available at higher pressures. The facility will be capable of simulating particular types and ranges of parameters, controls, and transients inherent in direct, indirect, and dual-cycle types of boiling water reactor power plants and will permit systematic investigation of the effects of these variables on the stability and performance characteristics of such reactor systems.

Power Distribution in Large Boiling Water Reactors

The distribution of steam in a boiling water reactor is not uniform. Radially, the steam content is highest in the regions of highest power production, and axially, the steam content increases continuously in the direction of steam flow. These nonuniformities in steam content perturb flux and power distributions from those which would hold for a reactor of uniform composition. In general, the power level will be reduced in the regions of high steam content. Radially, the effect is to flatten the flux distributions, a generally desirable effect. Axially, the effect is to decrease the power level at the exit end of the core relative to that at the entrance. If this effect becomes large, it may have a serious effect upon the total power capability of the reactor.

In reactors which are small, or more precisely in which the axial buckling is high, the reactivity is quite sensitive to these changes in flux distribution since the changes usually cause important increases in neutron leakage. Conversely it may be said that in such reactors large changes in flux distribution cannot be supported by the neutron balance in the chain reaction, or that the

flux distribution in such reactors is "stiff" and not subject to large variations unless the nonuniformities of composition are quite large. In EBWR, which is a relatively small reactor, the calculated perturbation of the flux distribution is a relatively minor one.⁴

For large boiling reactors the effect of non-uniform steam bubble density can be quite important. Detailed calculations for such a reactor have been carried out⁵ by an iteration technique in which a steam bubble distribution is assumed and the resultant power distribution is calculated, which, in turn, yields a corrected steam bubble distribution. The calculations are continued until a self-consistent power distribution is attained. The results of these calculations reveal several significant effects.

1. Boiling commences below the axial mid-point of the core so that the axial power distribution has a very large peak to average, almost the same as if a bank of control rods were near the mid-point of the core.

2. The radial and axial power distributions are coupled so that the diffusion equation is no longer separable.

3. Auxiliary means are required to increase the heat transfer in the upper part of the core. The methods being considered are stagnant water in the upper core region, rods programmed from the bottom of the core, and nonuniform fuel.

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Experimental Breeder Reactor

A series of papers were presented at the June meeting of the American Nuclear Society on the Experimental Breeder Reactor (EBR-I) meltdown,¹⁻⁴ describing the experiment, an analysis of the incident, the damage done to the core, and a description of the new core design for the reactor.

The experiment being performed was an attempt to measure transient temperature coefficients by measuring changes in reactivity of the reactor as the temperature of the fuel elements was increased. In order to obtain the temperature coefficient of the fuel alone, the coolant flow was shut off during the experiment. At the beginning of the experiment, the period meters were disconnected and the safety circuit trips were set above 1100 kw. The reactor was placed on a short positive period, and the fuel temperature was allowed to rise. On the final run of the series of experiments, the meltdown occurred due to a misunderstanding of the operating orders given for shutdown. The peak temperature recorded was 650°C. This was known to be low because of the lag time of the thermocouples. The maximum temperature is known to have been less than 1400°C, however, because the thermocouples in the core were found to be intact and their melting point is 1400°C. Peak power was estimated at between 15 and 30 Mw.

An analysis of the power vs. time data obtained in this experiment indicates that the decrease in reactivity as a function of time matched that to be obtained by the normal dropping of the external blanket. From this it was concluded that the shutdown was effected by dropping of the external blanket (actuated either by the over-power safety circuit or by the manual scram button) and not by core expansion.

The core meltdown necessitated the removal of the damaged core from the reactor. A temporary cave was constructed on top of the reactor to permit the removal of the core from the reactor as a unit. The core section was then cut from the handle section of the rods to remove it from the reactor assembly. This operation permitted the removal of approximately the two

outer rings of fuel elements from the core. The remainder was shipped to ANL as a single piece where it was disassembled in a high level cave. The pyrophoric hazard was largely removed during the disassembly operation by the use of a nitrogen atmosphere in the cave. The criticality hazard was handled by ensuring that the core could not be placed where it was adjacent to more than a single plane surface.

The disassembly showed that between 40 and 50 per cent of the volume of the core had melted and resolidified. The fused mass was in the shape of a prolate spheroid and contained three clearly defined density zones as a result of varying amounts of porosity. The central zone was found to be the most porous. The density of this zone was 2.5 g/cm³. The zone surrounding the upper half of the central zone had a density of 5.3 g/cm³, and the slump zone surrounding the lower half was found to have a density of 17.8 g/cm³ which was essentially the original density of the fuel. The higher density of the upper zone relative to that of the central zone was possibly due to the force exerted by the vaporization of the NaK in the center. The vaporization of the NaK was the most likely cause of the porosity in the mass, although the porosity could have been caused by the release of fission gases. Alloying in the molten zone between the fuel and structural materials was found to be complete. Later experiments showed that the spongelike structure in the core could be simulated by pouring molten eutectic of uranium and stainless steel into NaK.

A new core design has been completed to replace the core which was melted down. The design will permit the installation of either a rigid or loose core to study the effect of rod bowing on stability. Rod bowing is thought to be the most likely cause of the instability in the previous core. Valving has been incorporated into the design to permit either series or parallel flow through the core and blanket and throttling of parallel blanket flow. For the rigid core design, which will be installed first, the fuel and blanket rod design consists of coextruded U-Zr alloy rod with Zircaloy-2 clad. Wire spacer ribs are spot-welded on the outside of the clad.

An oscillating rod system and thorough thermocouple instrumentation have been designed for the power coefficient of reactivity tests to be run.

The Oxide Fueled Fast Breeder

Choice between oxide and metal as a fast reactor fuel is less straightforward than the corresponding choice in the case of water cooled reactors, where the oxide exhibits an important advantage in its chemical inertness to water. In the case of the fast reactor, it can be said that metallic fuel elements have the advantage of having been proved in the EBR-I reactor and are expected to give appreciably better physics performance because of their higher density. On the other hand, it is anticipated that oxide elements will be quite resistant to radiation damage and therefore will permit high burn-up. Oxide elements are expected to be cheap to fabricate, and the reprocessing of oxide fuel is somewhat simpler than the usual reprocessing procedures for metallic fuel. However, if pyrometallurgical reprocessing is developed for the metallic fuels, the latter advantage may not materialize.

The case for the oxide-fueled breeder was reviewed in three papers at the recent ANS meeting.⁵⁻⁷ Recent results on the development of plutonium oxide-uranium oxide elements were encouraging; the authors envisioned elements of this type which would run to 50 per cent burn-up of the initial plutonium. Multigroup reactor physics calculations were discussed which estimated the difference in breeding ratio between oxide- and metal-fueled reactors. The estimate was a loss of approximately 0.2 in breeding ratio in going from a metal core to an oxide core and a further loss of 0.05 if the metallic blanket were replaced by one using oxide. These losses were considered to have only a small economic

effect in a nuclear power economy which prices plutonium as a reactor fuel rather than as a weapons material.

Earlier reports on these investigations are contained in references 8 to 10.

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The organic moderated reactor is unusual in that it has a single feasibility problem—the dissociation of the organic under irradiation and high temperature—which has an overwhelming effect on its future usefulness. Since a reactor experiment is being prepared, there is a strong incentive for deferring interest in the reactor until the results of the experiment are known. A recently issued compilation¹⁻³ of information on organic moderator and coolant technology collects the information which is presently available and will probably be the definitive publication on this type of reactor until the reactor test has yielded results.

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GAS COOLED REACTORS

An Oak Ridge School of Reactor Technology (ORSORT) study of a gas cooled natural-uranium D₂O moderated power reactor, made in 1956, has recently been declassified.¹ The study treats nuclear characteristics, heat transfer, and power cycle, but no engineering design has been made. A fuel element is proposed which consists of a spiral-wound plate of zirconium-clad natural uranium. Such spirals are stacked in vertical process tubes. Cool inlet gas flows up an annulus between the process tube and the outer turn of the spiral, spirals inward between the closely spaced consecutive turns of the spiral, and is collected from the inner coil of the spiral. This design permits a short heat-transfer flow path while utilizing the full length of the reactor core for fuel loading. The process tube is a pressure tube which maintains the helium coolant at a pressure of 500 psi, whereas the relatively cool moderator is held at a pressure of ~200 psi. The reactor core is 20 ft in

diameter and 20 ft high and has a heat output of 1000 Mw. The coolant inlet temperature is 350°F and the outlet temperature 1000°F; the maximum fuel temperature is 1112°F. The conversion ratio is 0.70, giving an estimated fuel lifetime of 6000 Mwd/ton with natural uranium. The rather good heat-transfer performance, which makes possible the outlet temperature of 1000°F without exceeding the temperature of the uranium phase change, is dependent on the feasibility of the proposed fuel element design. No experimental data are given on the fuel element.

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The release of information¹ regarding the Aircraft Reactor Experiment (ARE), built at ORNL and operated in 1954, has aroused considerable interest in the use of molten fluorides in reactors. The authors point out that the fluorides of the alkali metals, beryllium, and zirconium mixed with the fluorides of uranium and thorium satisfy three major requirements for a desirable fluid fuel: large range of solubility for thorium and uranium, low pressure requirements, and the lack of radiolytic gas production. The nuclear properties and stability under high temperatures are also good, and the heat-transfer properties compare favorably with those of water. Certain disadvantages are also apparent: the corrosive nature of the fluorides and the high initial melting points. Beyond these problems, it can only be recognized at this time that the molten-fluoride reactor has the same general problems as other homogeneous reactors—maintenance difficulties and the necessity of absolute leaktightness and extreme reliability of components—and that while it may alter these problems it is not certain whether they are aggravated or relieved. The promise of high-temperature operation and the resulting high thermal efficiency make the system attractive provided that the homogeneous reactor problems can be economically solved and that an adequate and reliable system can be developed.

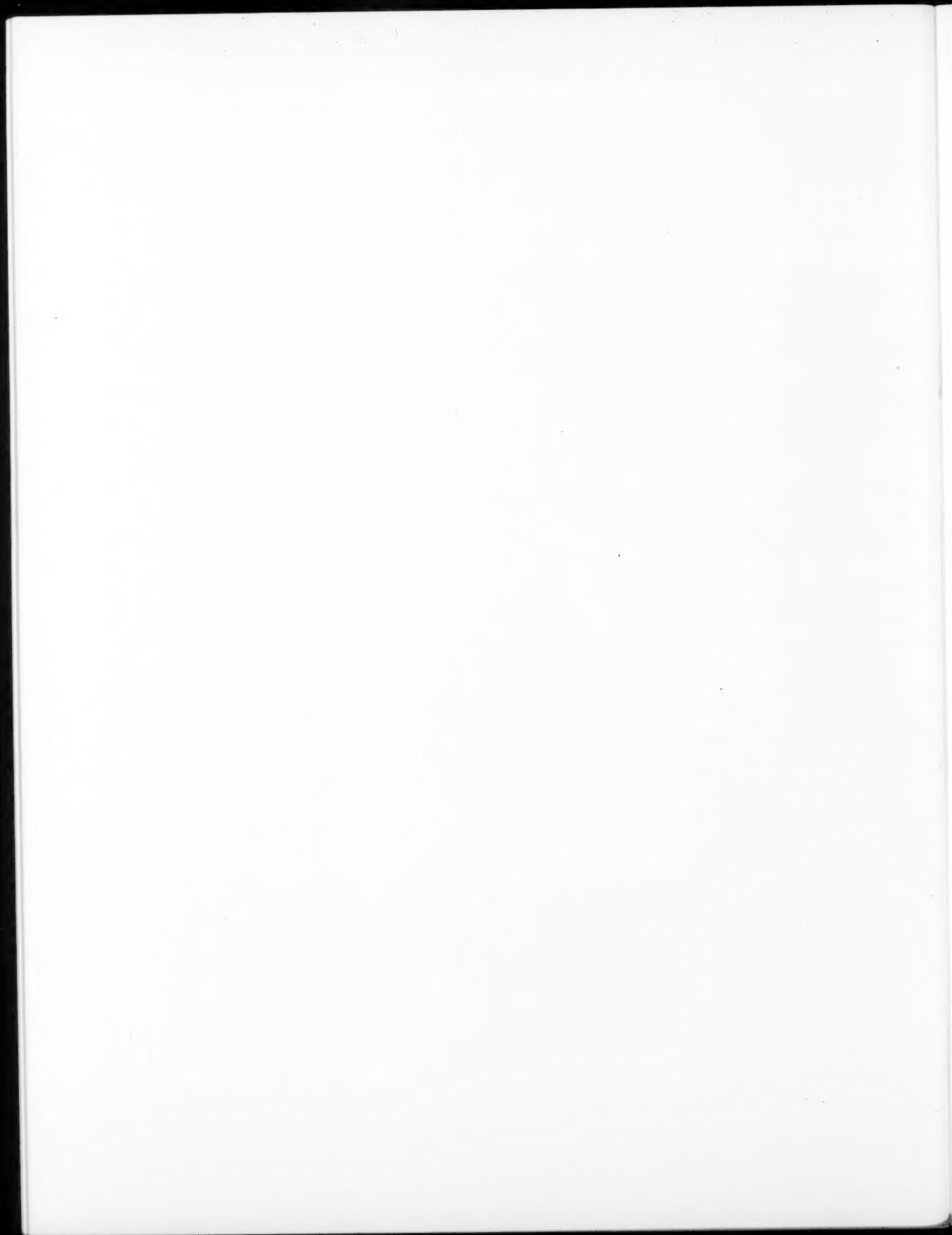
Conversion in the molten fluoride reactor is conceived to take place in an external blanket.

The possibility of fluoride mixtures containing ThF_4 represents some simplification over the case of the liquid-metal fueled reactor, in which a slurry is visualized for the blanket. The problem of a low-absorption container for the core still exists however. For applications in the near future, the reactor is proposed¹ as a straight U^{235} burner. It is pointed out that if full burn-up of the U^{235} can be realized, and if the high-temperature performance of the reactor makes possible a net efficiency of 35 per cent, the fuel burn-up cost for such a system would be between 2 and 3 mills/kw-hr.

It is interesting to note that considerable Russian work has been done on the chemistry of the fused-salt systems based on thorium fluoride.^{2,3}

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